

**Implementation and commissioning of the
phase-imaging ion-cyclotron-resonance method
and mass measurements of exotic copper isotopes
with ISOLTRAP**

DISSERTATION

zur Erlangung des Grades

Doktor rerum naturalium

(Dr. rer. nat.)

vorgelegt

der Fakultät Mathematik und Naturwissenschaften
der Technischen Universität Dresden

von

Dipl.-Phys. Andree Welker

geboren am 13.02.1986 in Bad Kreuznach

Eingereicht am 19.03.2018

Gutachter: Prof. Dr. Kai Zuber

Prof. Dr. Klaus Blaum

Tag der Verteidigung: 25.06.2018



Gewidmet

meinen Eltern

Andrea und Fred Welker

und

meiner Lebensgefährtin

Karmen Džeko

Zusammenfassung

Ein wesentlicher Bestandteil moderner Kernphysik ist die Beschreibung der Zusammensetzung des Atomkerns durch experimentelle und theoretische Anwendungen. Mit lediglich zwei Komponenten, den Protonen und Neutronen, trägt dieser eine Vielzahl von Informationen, welche über dessen Struktur zugänglich werden. Eine dieser Parameter, der Aufschluss über die Struktur geben kann, ist die Masse des Atomkerns, mit der anhand der Bindungsenergie der einzelnen Konstituenten, Schalenabschlüsse, und damit die Stabilitäten sowie die Wahrscheinlichkeiten solcher Strukturen erklärt werden können. Diese Informationen geben unter anderem Aufschluss darüber, wie in einer Verschmelzung eines Doppelneutronensternsystems oder einer Supernova, die Elemente, wie etwa Kupfer, Silber und Gold überhaupt erst entstehen können.

Diese Dissertation gibt erste Einblicke in die neue phasenbildgebende Ionenzyklotronresonanztechnik an ISOLTRAP, die ein noch höheres Auflösungsvermögen von Isomeren- und Grundzuständen in einem kürzeren Messzeitraum als bei bisher verwendete Techniken ermöglicht. Somit ist es dieser neuen Technik erlaubt kurzlebige Isotope zu vermessen als bisher. Den Einbau sowie die Charakterisierung in direkter Gegenüberstellung der alten Flugzeitmessmethode werden evaluiert. Darüber hinaus werden Untersuchungen des Elektroneneinfangs, des Paares ^{202}Pb - ^{202}Tl , vorgestellt, mit denen gezeigt wird, ob sie der präziseren Bestimmung der oberen Neutrino-Massengrenze dienen können. Die hochpräzise vermessenen Massen entlang der neutronenreichen Kupferisotope $^{75-79}\text{Cu}$, welche den Neutronenschalenabschluss $N = 50$ erreichen, werden mit ihrer Kernstruktur im Vergleich zu Berechnungen eines Schalenmodells verwendet, um das Verhalten des wichtigen Wartepunktnuklids ^{78}Ni zu interpretieren. Abschließend werden die ersten Massenmessungen der Isotope $^{123g,m}\text{Cd}$ und $^{127g,m}\text{Cd}$ unter Verwendung des phasenbildgebende Verfahrens vorgestellt, welche an ISOLTRAP vermessen und mit bestehenden Werten anderer Einrichtungen validiert wurden. Die Dissertation schließt mit einer Zusammenfassung und einem Ausblick auf weitere Verbesserungsmöglichkeiten der neuen Phasenbildtechnik sowie einem kurzen Überblick über weitere physikalisch relevante Kandidaten im Bereich der Massenspektroskopie.

Abstract

An essential component of modern nuclear physics is the description of the composition of the atomic nucleus through experimental and theoretical applications. With only two components, the protons and neutrons, it carries a lot of information which is accessible via its structure. One of these parameters, which can provide information about the structure, is the mass of the atomic nucleus, with which on the basis of the binding energy of the individual constituents, shell-closures, and thus the stability and probabilities of such structures can be explained. Among other things, this information provides the explanation on how elements such as copper, silver and gold can be formed in the first place in a double neutron star merger or in a supernova.

This dissertation gives first insights into the new phase-imaging ion-cyclotron-resonance technique at ISOLTRAP, which enables an even higher resolving power of isomeric- and ground-states in a shorter measuring period than with previously used techniques. Thus, this new technique allows to measure more short-lived isotopes than before. The installation as well as the characterization in direct comparison of the old time-of-flight measurement method are evaluated. Furthermore, studies of the electron capture, of the pair ^{202}Pb - ^{202}Tl , are presented, with which it is shown whether they can serve the more precise upper neutrino mass limit determination. The high-precision measured masses along the neutron-rich copper isotopes $^{75-79}\text{Cu}$, which reach the neutron shell-closure $N = 50$, are used with their nucleon structure in comparison to a calculation of a shell model to interpret the behavior of the important waiting point nuclide ^{78}Ni . Finally, the first mass measurements of the isotopes $^{123g,m}\text{Cd}$ and $^{127g,m}\text{Cd}$ are presented using the phase imaging method, which were measured at ISOLTRAP and validated with existing values of other facilities. The dissertation concludes with a summary and an outlook on further improvement possibilities of the new phase imaging technique as well as a brief overview of further relevant candidates with respect to mass spectroscopy.

Contents

Introduction	1
1 Basic notions of nuclear physics	5
1.1 Specific observables of a nucleus	5
1.1.1 Decay-modes	5
1.2 The quantum numbers	7
1.3 The nucleus' mass	9
1.4 Nuclear Mass Models	12
1.4.1 Liquid-drop model	12
1.4.2 Nuclear shell model	15
1.5 Finite-difference mass formulas	19
1.5.1 Separation Energies	19
1.5.2 The empirical pairing-gap	21
2 The ISOLTRAP experiment	23
2.1 The ISOLDE facility, a radioactive ion beam facility	23
2.2 The ISOLTRAP experiment	25
2.2.1 Radio-Frequency Quadrupole	27
2.2.2 Multi-Reflection Time-of-Flight Mass Separator	29
2.2.3 Introduction of Penning traps	32
2.2.4 Penning traps working principle	41
2.2.5 Preparation Penning trap	44
2.2.6 Precision Penning trap	48
2.2.7 Various ion detection techniques	49
3 Mass measurements with ToF-ICR	57
3.1 ^{202}Pb - ^{202}Tl a pair to determine the neutrino's upper mass limit? . . .	58
3.1.1 Measurement	59
3.1.2 Results	61
3.1.3 Conclusion	62
3.2 ^{79}Cu and its relevance for the determination of the ^{78}Ni 's mass surface	63
3.2.1 Measurement	64
3.2.2 Analysis	65
3.2.3 Results	68
3.2.4 Conclusion	73

4	Phase-Imaging Ion-Cyclotron-Resonance technique	75
4.1	Phase-Imaging Ion-Cyclotron-Resonance principles	76
4.1.1	I) Independent measurement method of ν_- and ν_+	77
4.1.2	II) Direct measurement method of ν_c	79
4.2	A direct comparison of the novel PI-ICR and the standard ToF-ICR technique	80
4.2.1	Initial spatial distribution	80
4.2.2	The resolving power in comparison	83
4.2.3	The precision of both techniques	84
4.3	Adjustment of ISOLTRAP for the PI-ICR technique	86
4.3.1	The delay-line position-sensitive detector	86
4.3.2	ISOLTRAP's PI-ICR data-acquisition system	89
4.3.3	A new ejection scheme & further changes for ISOLTRAP	90
4.4	PI-ICR tuning strategy for ISOLTRAP and systematic uncertainties	96
4.4.1	Minimizing the initial axial motion	96
4.4.2	Minimizing the initial magnetron motion and conversion admixture	97
4.4.3	Harmonizing the electric and magnetic trapping field	100
4.4.4	Residual gas interaction	105
4.4.5	Disruptive factors of the frequency to be measured	108
4.4.6	Inclinations and their impact on the frequencies	113
4.4.7	Interconversion of radial motions and the shift effect of ν_+	120
4.4.8	Search for additional accumulation time shifts	125
4.4.9	Summary of possible uncertainties at ISOLTRAP	127
5	Mass measurements with PI-ICR	129
5.1	$^{123,127}\text{Cd}$ ground states and their isomeric masses	129
5.1.1	Measurement	130
5.1.2	Results	131
5.1.3	Conclusion	133
6	Summary and Outlook	135
	Appendices	141
A.1	Simulated spatial ion distribution and its corresponding axial mean ToF	142
A.2	Preparation and precision Penning trap transport voltages	143
A.3	Lower trap and upper trap magnetic field	144
A.4	Upper trap capture delay and axial deflection	145
A.5	Upper trap axial motion	146
A.6	Mean ToF and the unlocked magnetron motion phase scan	147
A.7	Comparison between the resolving power of ToF-ICR and PI-ICR in case of ^{129}Cd	148
	Bibliography	154

List of Figures

1	The chart of nuclides by E. G. Segrè.	3
1.1	The binding energy per nucleon E_{bin}/A calculated with the liquid-drop model values of A. H. Wapstra.	13
1.2	The residual binding energy per nucleon after the reduction of the predicted value of the Bethe-Weizsäcker formula.	14
1.3	The figure represents the effective Hamiltonian $h_{lj}(r)$ with the corresponding sub-shell degeneracy of $2j + 1$	18
1.4	The experimental binding energies, as well as different applied mass filters for the potassium (K) isotopes.	20
2.1	The ISOLDE facility with its experiments.	24
2.2	A periodic table with the elements available as radioactive beams at ISOLDE.	25
2.3	The schematic representation of ISOLTRAP as the horizontal and the vertical beam line.	26
2.4	Schematic drawing of ISOLTRAP's radio-frequency quadrupole cooler and buncher.	28
2.5	ISOLTRAP's Multi-Reflection Time-of-Flight Mass-Separator (MR-ToF MS).	29
2.6	Schematic drawing of the cylindrical and hyperbolic Penning trap of ISOLTRAP with an additional plot to exemplify the three decoupled eigenmotions.	33
2.7	Diagram of the Landau level splitting described by harmonic oscillators for spin-less charged particles.	37
2.8	The hyperbolic Penning trap ring electrodes with a schematic sketch of the electrical field lines of the dipole and quadrupole excitation.	42
2.9	The dipole excitation on the magnetron motion frequency in relation to the ion motion phase.	43
2.10	The three decoupled eigenmotions of a charged trapped particle in an axial electrostatic quadrupole trapping potential, superimposed by an axial static magnetic potential.	46
2.11	The theoretical trajectory of $^{133}\text{Cs}^+$ in the preparation Penning trap.	48
2.12	The theoretical undamped radial motion trajectory of $^{133}\text{Cs}^+$ in the precision Penning trap with the applied quadrupole radio-frequency ω_c	49

2.13	The typical one-pulse and Ramsey type ToF-ICR technique for $^{133}\text{Cs}^+$ with an excitation time of $T_{ex} = 1200$ ms is shown.	51
2.14	Comparison of the mass measurements of ^{129}Cs by use of the one-pulse ToF-ICR, the two-pulse Ramsey and the PI-ICR technique with five measurements each.	54
2.15	Shown are the resulting mass-dependent shifts of ions available at ISOLTRAP for $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$	55
3.1	Shown is a ToF-ICR measurement of $^{202}\text{Tl}^+$ with an relatively long excitation of $T_{ex} = 3000$ ms and the measured cyclotron-frequency ratios with respect to $^{133}\text{Cs}^+$	60
3.2	Available copper yields obtained from the ISOLDE yield database. . .	64
3.3	One figure represents the ToF-ICR measurement of $^{78}\text{Cu}^+$, the other the MR-ToF MS spectrum of $^{79}\text{Cu}^+$ and isobaric species after $n_r = 1000$ revolutions.	65
3.4	Represented are the MR-ToF mass measurements of the isotope ^{75}Cu with four different scenarios, as well as the measurements of ^{77}Cu highlighted with different number of revolutions.	66
3.5	The JUN45 and the PFSDG-U shell-model valence spaces are compared to each other.	68
3.6	Experimental two-neutron separation energies along the nickel, copper and zinc chains compared to shell-model calculations.	69
3.7	Occupation levels from the PFSDG-U calculation given in total numbers of protons and neutrons along the copper chain for even and for odd and even N	71
3.8	Empirical two-neutron shell gap $\Delta_{2n}(Z, N_0)$ of neutron-magic nuclides and the shell gap for $\Delta_{2n}(Z, N_0 - 2)$	72
4.1	The phase-imaging projection of the ions motion in the Penning trap towards the position-sensitive detector for $^{85}\text{Rb}^+$ ions, on a 0.5 mm magnetron radius.	76
4.2	The PI-ICR independent measurement excitation-pulse scheme, for the magnetron and the reduced cyclotron frequencies.	77
4.3	The PI-ICR direct measurement excitation-pulse scheme, for the magnetron and the reduced cyclotron frequencies.	79
4.4	Comparison of different magnetron radii in the trap using the ToF-ICR technique and projected onto the detector by use of the PI-ICR technique.	82
4.5	The theoretically attainable mass resolving powers of different techniques used at ISOLTRAP.	85
4.6	The position-sensitive multi-channel plate detector (PS-MCP), with its delay-lines is illustrated in a rendered CAD-drawing and superimposed with its corresponding signal response.	87
4.7	The precision Penning trap tower is illustrated and superimposed by the magnetic field strength trend from the trap towards the detector.	91

4.8	Schematic overview of the new precision Penning trap control cycle for the ToF-ICR and the PI-ICR technique.	92
4.9	The illustration shows the electrical field disturbances generated by the old optical elements and the old detection devices.	94
4.10	The measured projections of the different motions in PI-ICR for the isotope $^{85}\text{Rb}^+$ are shown.	95
4.11	The picture represents the mean ToF distribution of the ions in conjunction with different axial energies, correlated as a function of different capture times of ISOLTRAP's precision Penning trap.	96
4.12	Shown are the results of the ToF-ICR cyclotron conversion procedure at different ion magnetron motion phases with their corresponding ion ToF, as well as a scan of the magnetron amplitude in order to obtain the smallest initial magnetron motion.	98
4.13	Presented is the quadrupole conversion amplitude scan with its corresponding mean time-of-flight behavior of the ions.	100
4.14	Demonstrated are the effects of different z^2 -shim compensation coil current values and their effect on the magnetic field homogeneity within a certain trap volume using the ToF-ICR and the PI-ICR technique.	101
4.15	The harmonization of the electrostatic trapping potential using the PI-ICR technique is shown.	105
4.16	Schematic drawing of the residual gas ion interaction and the behavior at the magnetron and reduced cyclotron motion radius.	107
4.17	The figure shows the behavior of the trapped ions, which interact with the remaining buffer gas in the precision Penning trap.	108
4.18	Illustrated are the magnetron and the reduced cyclotron frequency fluctuations recorded over 24 h at ISOLTRAP.	110
4.19	Schematic drawing of the ion motion projection to visualize displacement and inclination effects.	113
4.20	The obtained magnification factor P , measured at ISOLTRAP with respect to different drift electrode voltages, in comparison to a SIMION simulation.	115
4.21	Representation of the projection of the ion trajectory onto the position-sensitive detector and its effect on it with an inclined detector.	117
4.22	A novel method to estimate the detector or trap inclination with the symmetrical spot distribution on the position-sensitive detector in combination with the time-of-flight distribution.	118
4.23	Visualization of the admixture of motions and the ions which spiral along the magnetic field lines compared to simulations.	124
5.1	The separation of the $11/2^-$ isomeric-state from the $3/2^+$ ground-state of $^{127}\text{Cd}^+$ with an accumulation time of $t_{\text{accu}} = 150$ ms.	131
5.2	The recorded accumulation time trigger events for the FPGA card in case of an accumulation time $t_{\text{accu}} = 200$ ms.	132

A.3	The figure represents the transfer electrodes between both Penning traps of ISOLTRAP and their corresponding measured and simulated magnetic field strength along the transfer axis.	144
A.4	The figure shows the simulated ion axial deflections, resulting from different upper trap capture times.	145
A.5	The illustration shows a simulation that demonstrates the possibility of investigating the axial motion of the ions within the trap by projecting ions to different axial motion phases onto the position-sensitive detector.	146
A.6	The figure shows the effects of the axial motion on the mean ion time-of-flight.	147
A.7	The measurement of $^{129}\text{Cd}^+$ in 2014 with ToF-ICR and in 2017 with PI-ICR in direct comparison of the resolving power of the ground- and isomeric-state within an excitation, respectively, accumulation time of 100 ms.	148

List of Tables

1.1	Overview of four atomic physics quantum states.	8
2.1	Typical ion motion frequencies achieved with the ISOLTRAP's precision Penning trap.	36
2.2	Typical mass resolving power $\mathfrak{R}_{ToF-ICR}$, at the precision Penning trap of ISOLTRAP.	52
3.1	Cyclotron-frequency ratios of $^{202}\text{Pb}^+$, with respect to $^{133}\text{Cs}^+$ and $^{202}\text{Tl}^+$, with respect to $^{181}\text{Ta}^{16}\text{O}^+$, as well as $^{203}\text{Tl}^+$	61
3.2	Presented are the properties of ^{202}Pb , the spin, half-life and the electron binding energies of the daughter nuclide ^{202}Tl	61
3.3	Shown are the frequency ratios ($r = \nu_{c,ref}/\nu_{c,IOI}$), time-of-flight ratios (C_{ToF}) and resulting mass excesses (ME_{exp}) of the copper isotopes measured.	67
4.1	Specifications of the delay-line anode detector DLD40 from RoentDek GmbH.	86
4.2	Given is an overview of state-of-the-art position-sensitive detector concepts for efficient ion-detection.	88
4.3	Highlighted are the voltage settings for the precision Penning trap extraction and ion optics at ISOLTRAP, used for ToF-ICR and PI-ICR in comparison for the new and the previous extraction schemes.	90
4.4	Typical systematic shifts of the free cyclotron frequency $\delta\nu_c/\nu_c$ for the PI-ICR detection technique at ISOLTRAP with $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$ in comparison to SHIPTRAP with $^{133}\text{Cs}^+$	127
5.1	Measured frequency ratios, as well as the experimentally determined mass-excess ME_{exp} values of $^{123g,m}\text{Cd}$ and $^{127g,m}\text{Cd}$	132
A.1	Simulated projection of ions of mass $A = 85$ and a charge of $q = +1$, at the reduced cyclotron motion compared to different axial amplitudes.	142
A.2	The previous and the new, less mass-dependent voltages for the transfer electrodes between the preparation Penning trap and the precision Penning trap are shown.	143

Introduction

Our universe evolved, described with the prevailing cosmological standard-model (Λ -cold-dark-matter model), in a big bang. According to the current scientific knowledge, the big bang happened $13.799 \pm (0.026)$ Ga [Ade16] ago. This event was immediately followed by the Planck era, where the time and space was created, which lasted until 10^{-43} s after the big bang. The Grand Unified Theories (GUT-theories) [GG74], describe that the weak, strong and the electromagnetic force, were once unified. With 10^{-8} s, after the big bang, those forces, were decoupled from each other. The huge space expansion within 10^{-2} s afterwards inflated the universe [Gut81]. Consequently, this expansion caused a strong drop in temperature. The until then free evolving quarks, cluster to the first neutrons and protons by gluons and the interacting strong force, within the time span of 1 s - 3 min after the big bang [GM62]. It was the time, when the first free electrons and neutrinos started traveling through space. From now on in the primordial nucleosynthesis era, where neutrons and protons started to merge to H^- , He^{--} [ABG48] and d^- [Coo14], it should take 400.000 a [Ade16] before the universe under decoupling of photons from baryons gets transparent. With this important step the information of the evolving universe gets accessible, after it cooled down to ~ 3000 K [Pov17], for nowadays technology. The probability that an ion was able to capture an electron rose. Over the time the first gas clouds with the more and more dominant rising of the gravity were created followed by the first pre-galaxy's and quasars which were created in between 400.000 a - 1 billion a. The background radiation cooled down to 2.7255 ± 0.0006 K [Fix09] in the time-span of 1 billion a - 3 billion a. Now the possibilities were given to form the galaxies and stars of today within the time of 3 billion a - today after the big bang, with the creation of our solar system, the earth and the human beings. The first written record, that humans developed such a philosophy that an internal structure of matter exists, started already 2400 years ago. The Greek Democritus of Abdera contemplated 420 b.c. that atoms are not perceptible in analogy with our senses and therefore we report properties that the atoms don't really possess, like colors and tastes.

- νόμῳ γὰρ χιὴ, νόμῳ πικρόν, νόμῳ γλυκύ, ἔτεῃ δ' τὸ ἄτομον καὶ κενόν. -
It only seems a thing has a color, only apparently it is sweet or bitter; in reality
there are only atoms in an empty space.

(DK 68 B 125)[Her22]

A few millennia should have passed since then, until H. Becquerel¹ discovered at the end of the 19th century, the spontaneous emission of radiation from uranium compounds [Bec96]. The origin of this radiation was found later to be the uranium atom itself by G. Bémont, M. and P. Curie¹ [MCB98], during the studies of elements with a similar behavioral pattern, which was from then on coined as radioactivity. Soon it was clear that the atom is dividable and consists of electrons, which were discovered by J. J. Thomson² [Tho97] during the studies with the cathode radiation. With the radiative materials E. Rutherford and P. Villard classified three different types of radiation as α , β [Rut99] and γ [Rut03] by bombarding different materials, as well as the studies about the ionization of the air. Some years later, it was shown by E. Rutherford, H. Geiger and E. Marsden that penetrating a thin gold foil with α -particles under different angles revealed that the atom itself consists of a small and dense nucleus and an electron cloud around it [HG09; Rut11; HG13].

The first milestone to the branch of nuclear physics was made and the outstanding question of the internal structure of the nucleus was solved with the discovery of E. Rutherford's hypothetical neutral particle [Rut20], the neutron, as it was called by J. Chadwick³ [Cha32]. Thus, the substantial key knowledge came that an atom contains a certain amount of charged protons Z and electrically uncharged neutrons N , wherein the relation to the atomic number A evolves to $A = Z + N$. Using Z and N as parameters, C. F. Weizsäcker [Wei35] and H. Bethe [BB36] developed the first semi-empirical mass equation, the so called liquid drop model. The model and how it derives the binding energy E_{bin} , will be discussed beside other models in Section 1.4. During the following years more and more nuclides were discovered foremost by F. Soddy⁴ and F.W. Aston⁵ [Ast20; F.A20]. These continuous discoveries have been made possible by innovations such as the mass spectrograph of W. Wien [Wie98] and J. J. Thomson [Tho13], which uses the ability to deflect charged particles of a beam according to their mass-over-charge ratios m/q , in combination with electric and magnetic fields. This cornerstone of mass spectroscopy (MS) will be discussed more in detail in Section 2.2.2. Also big contributions are related to the developments of tools for the production of radioisotopes like the linear accelerator from R. Wideröe [Wid28] and the cyclotron from E. O. Lawrence⁶ [Law30; LL32], with which it is until today possible to discover additional exotic nuclides.

The more knowledge is obtained, the more information has to be correlated and interpreted in a correct, coherent and physical way, to find the roots of their natural origin. Therefore, the idea to design a chart of nuclides in a clear representative and

¹1903 Nobel Prize in physics for the discovery and description of the radioactivity.

²1906 Nobel Prize in physics for the theoretical and experimental investigations on the conduction of electricity by gases.

³1935 Nobel Prize in physics for the discovery of the neutron.

⁴1921 Nobel Prize in chemistry for chemistry behavior of different radioisotopes and their nature.

⁵1922 Nobel Prize in chemistry for the discovery of the isotopy of numerous stable elements and the whole number rule.

⁶1939 Nobel Prize in physics for the invention and development of the cyclotron and the obtained results.

logic way, with the protons organized along the ordinate (y -axis) and the neutrons along the abscissa (x -axis), started with G. Fea and E. G. Segrè [Fea35] and is shown in Fig. 1.

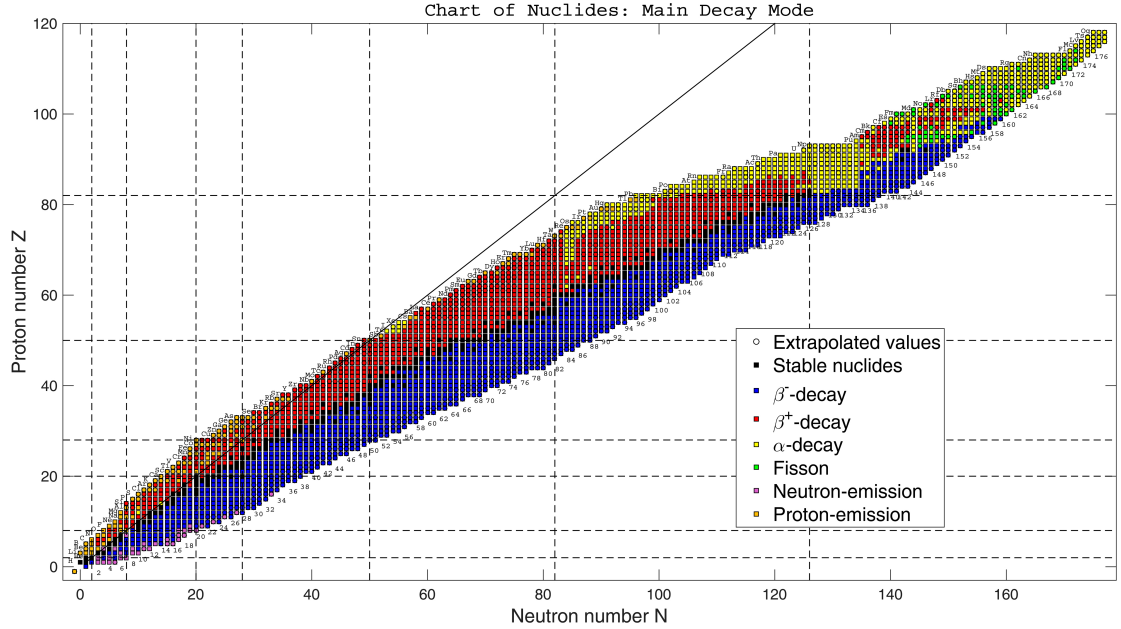


Figure 1: The chart of nuclides by E. G. Segrè represents the neutron number N given as a function of the proton number Z . Here the color code highlights the most abundant decay mode for each isotope, separated in α -, β^- , β^+ , fission and evaporation processes of protons and neutrons [Aud17]. The stable nuclides in the center of the chart labeled with a black color, compared to the non-stable ones depicted in the inset.

The chart contains, inter alia, the decay mode, the mass number, half-lives and energies of the most abundant decays. The chart of nuclides contrasts with the periodic table [Men69], which only maps their chemical behavior, since isotopes of the same element do not differ chemically, having the same electron orbit configuration.

During the studies around 1960, E. Wigner⁷ and M. G. Mayer⁷ [MJ55] raised the idea of a nuclear shell structure, in analogy to the atomic one. It was realized that some of the nuclei are extremely stable at certain magic nucleon numbers $\{Z, N\} = \{2, 8, 20, 28, 50, 82, \dots\}$. This model will be described in more detail in Section 1.4. In order to investigate the phenomena of the nuclear shell-structure, modern nuclear physics tools are able to probe the achievable observables of a nucleus. To illustrate just a few, the binding energy E_{bin} and the resulting mass m of a nuclide. The root mean-square charge radii $\langle r^2 \rangle$, to know how big the nucleus itself is, as well as the electric En and magnetic Mn ($n \in \mathbb{N}$) transitions between nuclear quantum states, which can be used to indicate how deformed a nucleus is. These information are used to verify theories, models, as well as to interpret their causality.

The binding energy E_{bin} , as an important observable, quantifies the strength of the

⁷1963 Nobel Prize for the development of the shell model and theory.

binding between each nucleon. This observable can be determined by use of, e.g. the high-precision mass spectrometer ISOLTRAP, at which the following thesis was written. The precise knowledge of the binding energy serves as a puzzle piece and opens the access to describe, amongst others, the formation of the universe, as well as the chemical element production mechanisms in connection with stars, in turn, the paths nature follows.

This work focuses on the implementation of a position-sensitive detector at ISOLTRAP, with which the experiment is able to study the nuclides with the novel phase-imaging ion-cyclotron-resonance (PI-ICR) technique. This novel technique was first established by the Max-Planck-Institute for Nuclear Physics in Heidelberg [Eit09], later extended and applied at the SHIPTRAP facility at GSI Darmstadt in Germany [Eli13; Eli14]. PI-ICR allows to measure masses with an even higher precision in a shorter measurement-time. The gain in resolving power which is achieved by the technique, gives the ability to study isomers with an even smaller excitation energy of a few hundred keV, compared to previous techniques. A brief overview of the basics in nuclear physics, relevant for this work, is given in Chapter 1. Subsequently, the ion production of the measured isotopes, the experimental setup and the used measurement techniques at the ISOLTRAP experiment, as well as the position-sensitive detector working principles are introduced in detail in Chapter 2. Furthermore, the evaluation of the electron capture (EC) Q -value measurement of $^{202}\text{Pb} \rightarrow ^{202}\text{Tl}$, to check if this pair of nuclides are good candidates for the determination of a more precise upper neutrino-mass limit, will be discussed in Section 3.1. The detailed investigation in the mass determination of the neutron-rich copper (Cu, $Z = 29$) isotopes $^{75-79}\text{Cu}$ is subject of Section 3.2. The section clarifies the $N = 50$ neutron closed-shell ^{79}Cu as a sensitive probe for shell-structure studies only one proton above the potentially doubly-magic nuclide ^{78}Ni . The two measurements, which were determined with the help of the Time-of-Flight Ion-Cyclotron-Resonance (ToF-ICR) technique, are followed by the characterization of ISOLTRAP's precision Penning trap by use of the PI-ICR technique in Chapter 4. Concluding with the first results of the proof of principle studies of the PI-ICR method at ISOLTRAP with the isomeric separation of $^{123g,123m}\text{Cd}$ and $^{127g,127m}\text{Cd}$, presented in Section 5.1. Details on the data analysis, the physics impact, as well as the results for each measured nuclide are conducted within each chapter. The dissertation concludes with a summary and an outlook on possible technical improvements at ISOLTRAP and physical aspects.

Chapter 1

Basic notions of nuclear physics

The power of high-precision mass measurements which provides the meticulous information of the mass, with relative uncertainties of $\delta m/m \leq 10^{-7}$ can be used to validate theories, models and predictions in the terrain of, e.g., weak-interactions, nuclear structure or nuclear astrophysics. Mass data is a key feature to derive input parameters for empirical mass formulas and to predict masses of unmeasured nuclides [LPT03]. Up to now, more than 3000 nuclides have been discovered [Wan17], which were determined from different and complementary experiments. Conclusions of a consistent picture of a nucleus or an isotopic chain can be drawn if its properties, such as the total angular momentum, the parity, the radioactive decay modes, the nuclear excited states and the masses are available. The access to those information is getting more and more difficult, the more exotic a nucleus becomes, i.e. nuclides far away from the stable and long-lived isotopes. The challenging and limiting factors are, e.g., the half-lives and the production rates which can be smaller than one nuclide per second [col16]. The limits of the nuclear 'landscape' which are represented in the neutron- and proton drip-line [Erl12], define how many neutrons and protons can be bound at maximum in a nucleus. The progress in this fields during the recent years, is to be able to extrapolate to those mass regions.

In the following, a brief summary of models and observables of a nucleus which are of importance for the masses investigated within this thesis, will be given. A more detailed overview can be found for example in [Ken87].

1.1 Specific observables of a nucleus

1.1.1 Decay-modes

More than 2500 nuclides of the known isotopes are unstable [Aud17]. The criteria to be able to measure exotic nuclides is to understand the decay modes of the specific nucleus, as well as its half-life. Such a criteria can be an estimation of the probability, if a measurement, with nowadays technologies, is feasible in first place. The decay gives also access to nuclides which might not be accessible immediately from the

point of the suppressed production mechanism or the short half-life, e.g., that the ion cannot be transported in time over the whole distance from the creation point up to the spectrometer, before it decays [Asc16]. But it also rises the possibility that the excited nucleus (isomeric state) can be reached if a mother nuclide decays into a metastable state of the daughter nucleus. A radioactive decay is a stochastic process by which the nucleus of an unstable atom loses energy by emitting radiation.

This decay process is described as:

$$n(t) = n_0 \cdot e^{-\frac{t \cdot \ln 2}{T_{1/2}}}. \quad (1.1)$$

Here n_0 represents the initial quantity of the mother nuclide, where $T_{1/2}$ is the time, in which on average half of the nuclides have been decayed. The main radiation emitting branches are the α -, cluster-, β^- -, β^+ -decays, electron capture (EC) and the internal transitions (IT), where the latter emits a photon γ . In the following X describes the mother nucleus, wherein X^* is the excited form of the mother nucleus and Y the daughter under a release of its excess energy ΔE .

- α - and cluster-decay:

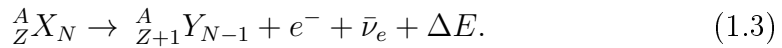
It is the definition of the tunneling process, if the nucleus emits an ^4He - or a bigger nuclei, preferably ^{14}C , ^{24}Ne , ^{25}Ne and ^{28}Mg , respectively. The following example shows the α -decay:



This process is more likely to happen for heavy nuclei with $Z > 83$, where the Coulomb repulsion of the protons increases faster than the attractive nuclear force. In the chart of nuclides, shown in Fig. 1, the nuclides decaying preferably by α -decay, are highlighted in yellow.

- β^- -decay:

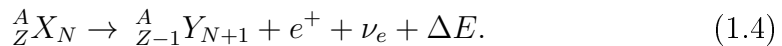
An unstable atomic nucleus with an excess of neutrons tend to undergo the β^- -decay, where a neutron is converted into a proton by releasing an electron e^- and an electron-antineutrino $\bar{\nu}_e$:



The color-code in the chart of nuclides, to highlight the β^- -decay in Fig. 1, is blue.

- β^+ -decay:

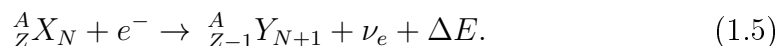
Respectively to the previous decay, the β^+ -decay is a process, of an unstable atomic nucleus with a deficiency of neutrons, within this process a proton is converted into a neutron by emitting a positron e^+ and an electron-neutrino ν_e :



The nuclides which preferably decay by the β^+ -decay channel, as shown in Fig. 1, are marked in red.

- EC:

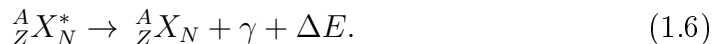
The electron capture is a competing process to the β^+ -decay¹, in which the neutron-deficient nucleus of an electrically neutral atom absorbs an inner atomic electron, usually from the K or L electron shell. This process thereby changes a proton into a neutron under simultaneous emission of an electron-neutrino. The EC is typically followed by emitting X-rays with a discrete fingerprint γ -energy, which occurs due to refilling of the electron hole from higher electron orbitals:



Both β -decays, as well as the EC are mediated by the weak force which changes their quark flavor through a gauge W -Boson. As an example for the β^- -decay, a down-quark in the neutron (u,d,d) is converted under emission of the e^- and an $\bar{\nu}_e$ with the interaction of a W^- -Boson into an up-quark. In this process the neutron (u,d,d) is converted into a proton (u,d,u). As an example the EC of the pair ${}^{202}\text{Pb}$ - ${}^{202}\text{Tl}$ and its relevance for the determination of the neutrino upper mass limit is described in Section 3.1.

- IT:

The internal transition and the reaction of an excited nucleus X^* is more a form of a de-excitation by emitting a γ than transforming itself into another element.



To get a deeper understanding of other possible decay modes the reader is referred to a have a look into [Pf2] which gives a well portrayed survey.

1.2 The quantum numbers

Many properties of nuclei can be reasonably well described by the simple shell model which resembles the shell model in atoms. One of these properties are the quantum numbers, which follow in the atomic cases as in some of the nuclides the Pauli exclusion principle [Pau25]. This principle describes that the total wave function for two identical fermions (particles with half-integer spin) is antisymmetric with respect to exchange of the particles. This means that two identical fermions cannot occupy the same quantum state simultaneously. Fermions include elementary particles such as quarks, electrons and neutrinos. Additionally, protons and neutrons called baryons

¹The decay channel of β^+ is only feasible, if the mass of the parent nucleus exceeds the mass of the daughter nucleus by at least two electron masses $2m_e = 1.022$ MeV. However, if the energy is less than 1.022 MeV the EC is the favored process.

(subatomic particles composed from three quarks) also have half-integer spin. A direct consequence of this is, e.g., ${}^3\text{He}$ with half-integer spin can be treated as a fermion, following the Fermi-Dirac-statistic, where ${}^4\text{He}$ with its full-integer spin, behaves as a bosonic system which follows the Bose-Einstein-statistics, therefore, it does not underlay the Pauli exclusion principle.

How the following atomic quantum states in Table 1.1 are determined, can be read in [Ken87].

Table 1.1: In the atomic physics the following four quantum states exists. Fermions (with half-integer value) follow the Pauli exclusion principle, whereas bosons (with full-integer values) due not. Electrons, protons and neutrons which are subject of fermions, have to be different in at least one quantum state. To describe the dominant effect of the spin-orbit coupling in a nucleus the quantum number (q.n.) m_j is additionally introduced in the following.

name	symbol	orbital description	range of values
principal q.n.	n	shell	$1 \leq n$
azimuthal q.n. (angular momentum)	l	subshell orbitals (s,p,d,f,g,...)	$0 \leq l \leq n-1$
magnetic q.n. (angular momentum projection)	m_l	energy shift (subshell's shape orientation)	$-l \leq m_l \leq l$
spin projection q.n.	m_s	electron spin $\pm\frac{1}{2}$	$-s \leq m_s \leq s$
angular momenta correlation	m_j	spin-orbit splitting $j = l+s$	$m_j = 2j+1$

Around 1960, E. Wigner and M. Goeppert-Mayer [MJ55] raised the idea of a nuclear shell structure, in analogy to the atomic one. The aspect to create such a model was that the central potential alone does not reproduce the behavior of the experimentally observed single-particle energies in the mean field. They also realized that the observed energies bunch into groups, or orbitals, similarly to the atomic case. These groups, separated by a distinct energy gap are called magic numbers. Further details will be described in Section 1.4. To reproduce these numbers theoretically, the spin-orbit² interaction has to be added to the mean-field potential. Due to the internal quark structure of the nucleons (neutrons n and protons p) an orbital angular momentum l , as well as a spin $s = \frac{1}{2}$, due to the fermion character, is present outside the nucleus. The wave function of the particles in the nuclear potential can be divided into two parts: a radial $R_{n,l}(r)$ which only relies upon the radius, and a part $Y_l^m(\Theta, \phi)$ which only depends upon the orientation. Under this circumstances the total angular momentum of a nucleus composes to:

$$\mathbf{j} = |\mathbf{l} \pm \mathbf{s}| \quad (1.7)$$

$$\mathbf{I} = \mathbf{j}_n + \mathbf{j}_p \quad (1.8)$$

²The atomic spin-orbit has the origin from the electromagnetic interaction in the milli electronvolts (meV) scale, where the nuclear spin-orbit has one dominant origin from the single-particle energy differences, within a million electronvolts (MeV) scale.

1.3 The nucleus' mass

One of the most momentous parameter of the nucleus is the knowledge of its mass with highest precision, from which the binding-energy E_{bin} of each nucleon in a nucleus can be determined. As J. J. Thomson started with his idea to determine mass-over-charge relations of nuclides, using electromagnetic fields 120 years ago [Tho97], he set the cornerstone of the precision mass spectrometry. This goal lasts until today, getting more and more precise, to reveal the secrets of nature. In order to establish the fields of mass spectrometry, many new excelling inventions emerged in the last century, such as in 1918 by A. J. Dempster [Dem18], who enhanced the previous mass spectrometry technique by a factor of 100. The achromatic mass spectrograph [MH34], an until today lasting apparatus for abundance analysis, developed in 1934 by J. Mattauch and R. Herzog. Between the years 1946 W.E. Stephens [Ste46], 1955 W. C. Wiley and I. H. McLaren [WM55] and 1973 B. Mamyrin [Mam73] explored and improved the time-of-flight mass spectrometry (ToF-MS) with the ability of time based pulses, triggers, read-outs and shape discriminated signals due to faster electronics. In parallel a new innovation of a radio-frequency (rf) electric field ion trap, the Paul trap, by W. Paul and H. Steinwedel [PS53] and the static electric and magnetic field Penning trap, by H. Dehmelt [BG86; Deh90] were accomplished and paved the way for today's most accurate and versatile mass spectrometers.

The relation between the mass of an atom $m_{at}(Z, N)$ and the mass of a nucleus $m_{nuc}(Z, N)$, is determined by:

$$m_{at}(Z, N) = m_{nuc}(Z, N) + Z \cdot m_e - \frac{E_{el}(Z)}{c_0^2}. \quad (1.9)$$

This includes the mass of electrons $Z \cdot m_e$, as well as the electrons binding energy $E_{el}(Z)$. Determine the nucleus mass $m_{nuc}(Z, N)$ reveals the binding energy $E_{nuc}(Z, N)$ of a nucleus. According to Einstein's energy-mass equivalence, the binding energy $E_{nuc}(Z, N)$ corresponds exactly to the mass defect $\delta m(Z, N)$ by the interaction of its constituents:

$$\delta m(Z, N) = Z \cdot m_p + N \cdot m_n - m_{nuc}(Z, N) = \frac{E_{nuc}(Z, N)}{c_0^2}, \quad (1.10)$$

where m_p is the proton mass and m_n the neutron mass. Here, c_0 is determined as the constant of the speed of light in vacuum. The nuclear force is the main origin of the nuclear binding energy³, from the interaction among nucleons (protons and neutrons), it is the residual of the strong interaction which bounds the quarks in a nucleon. Both nucleons, neutrons and protons, are affected by the nuclear force almost identically. Since protons have charge $q = +1e$, they experience in addition an electric force that tends to push them apart⁴. For this reason the binding energy

³From now on, the binding energy of a nucleus is written as E_{bin} , if not explicitly mentioned.

⁴The short-range of the residual strong interaction acts over 2.5 fm with the direct neighboring nucleons, where the coulomb interaction acts over an infinite range.

acts as a specific sum of energies, reflecting all interactions in a nucleus. The precise knowledge of the binding energy leads, for example, to explain the nucleosynthesis process and the description of the cluster probability between the protons and neutrons forming nuclei up to a certain drip-line. Thus, the binding energy explains why the largest stars in the universe can only produce up to the masses ^{56}Fe and ^{58}Fe with their fusion processes. All heavier elements have to be produced in rougher environments like supernovae, twin-stars, neutron-star mergers and black-holes [OJM07; AGT07]. Around $A = N + Z = 60$ the maximum binding energy is reached, as an example the strongest bound nucleus with $E_{bin} = 8794.553(7)$ keV/A [Wan17] is ^{62}Ni ⁵. To develop the theories and models for those descriptions, theoretical physicists are still searching for unified microscopic models. Therefore, precision mass measurements to derive the binding energy are still essential and indispensable. A small outlook for the models and theories which are relevant to this thesis, will be described more in detail in Section 1.4.

The following comparison shows with continually improved mass precision, more fields in physics can be pursued and solved. For further discussions the relative mass uncertainty will be written as $\delta m/m$.

- $\delta m/m < 10^{-5}$: This region is sufficient for chemical compound analysis, e.g., chemistry, environmental science, forensic, life-science for pharmaceutical tests, as well as analysis of extraterrestrial materials in astrophysics, e.g., Titan's atmosphere [Nie05], one of Saturn's moons. Nowadays commercial mass spectrometer are relatively fast in the analysis process and reliable within this mass region [Hü15]. To perform the detailed nuclear structure analysis, a higher precision is still necessary.
- $\delta m/m < 10^{-6}$: With this precision the nuclear shell effects, like shell closures, shell quenching, regions of deformation, the drip lines, halos, δV_{pn} ⁶ the interaction strengths between the last two protons and the last two neutrons, become visible in vicinity of magic numbers, with binding energy uncertainties smaller than $\delta E_{bin} \leq 100$ keV. These information give access for theories in the astrophysical sector to describe, e.g., the r- and rp-process, to reproduce and explain the natural elemental abundances in the solar system [LPT03]. With this accuracy the shell gaps [Wie13; Ros15], as well as the separation energies which will be described in detail in Section 1.5, are attainable.
- $\delta m/m < 10^{-7}$: The interaction between neutrons and protons in different orbitals can be tracked with the differences in the binding energies, compared to neighboring isotopes [Nai12]. The information of the binding energies reveal the ability for theoretical physicists to study astrophysical waiting points

⁵Lighter nuclei are covering more the surface of a nuclei with this property they feel less of the strong force, whereas the Coulomb interaction reduces the binding energy in the case of heavier nuclei.

⁶Describes and explains the shell structure and orbit occupations near the Fermi surface.

[AW15], neutron star crusts [Wol13a] and X-ray bursts [Cyb16; SO17]. First low-lying isomers with an excitation energy of $E_{Ex} \leq 50$ keV can be determined [Web05].

- $\delta m/m < 10^{-8} - 10^{-9}$: The weak-interaction studies allow to test the unitarity of the Cabibbo-Kobayashi-Maskawa (CKM) quark-mixing matrix⁷ by determine the first element V_{ud} . Further tests can be obtained to test the conserved-vector-current (CVC) hypothesis [Ero13; Par16] which can be derived from the fT -value of the super-allowed $0^+ \rightarrow 0^+$ β -emitters [HT15].
- $\delta m/m < 10^{-10}$: The search for the neutrinoless double β -decay or neutrinoless double electron-capture processes will reveal, whether neutrinos are their own antiparticles (Majorana particles) which would violate the lepton-number conservation (Physics beyond the Standard Model). Experiments which deal with neutrinoless double β -decay studies are GERDA [Ago17], SNO+ [OOC11] and COBRA [Zub01], wherein experiments like γ -ray emission in ^{58}Ni [Leh16], ^{156}Dy [FT15] and GERDA with ^{36}Ar [Ago16], explore the field of the neutrinoless double electron-capture.
- $\delta m/m < 10^{-11}$: The ambitious and outstanding challenging venture to determine the neutrino mass is an open question of modern physics. One opportunity to reveal the mass of the electron neutrino ν_e and the electron antineutrino $\bar{\nu}_e$ is to perform a high-precision Q -value measurement of the β -emitters, e.g., ^3H at KATRIN [Mye15; PSB15], to measure the end-point energy of the β^- -decay spectrum, as well as the Q -value mass measurement of the decay ^3H to ^3He with an uncertainty of $\delta m/m = 10^{-11}$, by us of the THE-Penning trap [Die11]. In addition, the Q -value mass measurements of ^{187}Re [Nes14] and the electron-capture Q -value of ^{163}Ho [Eli15] are some of the approaches. The new measurements will determine a more precise upper limit of the electron neutrino ν_e , with a current limit of 225 eV at 95% C.L., which derived from a measurement of spectra from the inner bremsstrahlung of ^{163}Ho [SBB87], as well as the electron antineutrino $\bar{\nu}_e$ mass with an upper limit of 2 eV at 95% C.L., determined by a tritium decay experiment [OW08]. With this accuracy it is possible to compare the antimatter with the ordinary matter mass. For example measurements of the mass or the charge ratios, e.g., m_{e^-} [Koe15], m_{e^+} , m_p [Hei17], $m_{\bar{p}}$, $m_{\bar{p}}/m_e$ [Hor16] and $Q_{\bar{p}}/Q_p$ [Ulm15], are acting as strong tests of the CPT-theorem [DiS13]. With the obtained precision it is now possible to approach the field of quantum electrodynamics (QED) in highly-charged ions (separation of atomic states), a field which will be affected by the PEN-TATRAN [Rou12]. Within this region of precision the fundamental constants [Mye13], e.g. the fine structure constant $\alpha_{e.m.}$ which can be tested with the mass ratios of h/m_{Cs} , m_{Cs}/m_p and m_p/m_e . The aimed precision provides access to novel provisions of the standard units to test, e.g. the Avogadro

⁷The CKM-matrix describes the transformation between the mass eigenstates and the weak interaction eigenstates of the Standard Model quarks.

constant (N_A) and the Planck constant (h) with a perfect crystal sphere of silicon (m_{Si}) [And11], as well as with the watt balance [Sto11].

The description above represented the importance of high-accurate measured masses and how the precise information affects physical aspects. The state of the art procedures to measure the masses of exotic nuclides are, e.g., storage rings [Lit13], multi-reflection mass spectrometers (MR-ToF MS) [Wol13b] and Penning traps [BNW10; BDN13], the both latter will be explained in more detail in Section 2.2.2 and Section 2.2.3, respectively.

1.4 Nuclear Mass Models

To be able to describe the origin of the nuclear structure, a theory has to be developed which can reproduce the experimental derived values reliably. Until now the lack of theoretical knowledge about the origin of the underlying force between the nucleons and the complexity of the many-body problem hinders the success. To understand in which manner the nuclear structure evolves, several mass models were designed. Nowadays, input parameters for empirical mass formulas and the prediction of unmeasured masses [LPT03] can be achieved with relative mass uncertainties of $\delta m/m \leq 10^{-5}$ - 10^{-6} . The main contributions to describe the mass of a nucleus can be separated into two sections, the macroscopic sector, described in Section 1.4.1 and the microscopic sector reflected in Section 1.4.2. In the following the focus will be on those theories and models which are used within this thesis.

1.4.1 Liquid-drop model

The first framework which described the binding energies with an accuracy of 99% [MK02], was the semi-empirical mass formula, shown in Eq. (1.11), named after their discoverer in 1935, C. F. Weizsäcker [Wei35] and H. Bethe [BB36].

$$E_{bin}(Z, N) = a_V \cdot A - a_S \cdot A^{\frac{2}{3}} - a_C \cdot \frac{Z(Z-1)}{A^{\frac{1}{3}}} - a_A \cdot \frac{(N-Z)^2}{4 \cdot A} - \frac{a_\delta(A, Z)}{A^{\frac{1}{2}}}. \quad (1.11)$$

The formula is also well known as the liquid-drop model which considers the atomic core as an incompressible fluid of nucleons, initially proposed by G. Gamow in 1930 [Gam30], with a constant density. The liquid-drop model is the macroscopic sector approach in the description of the atomic nucleus. Due to the premises that the formula cannot describe the shell effects, the strength of describing the mass region starts around $A > 30$. As mentioned in Section 1.3, the maximum of the binding energy peaks around ^{62}Ni with $E_{bin}=8794.553(7)$ keV/A [Wan17], followed by the solar system more abundant nuclides ^{58}Fe and ^{56}Fe ⁸, as can be seen in the figure

⁸For comparison: Nuclides around $A = 60$ are 0.9% lighter than the sum of the mass of its constituents. In contrast, the mass defect in an hydrogen atom, is on the order of $10^{-6}\%$, due to the electromagnetic force between the proton and the electron.

1.1, where the nuclear binding energy per nucleon E_{bin}/A is plotted as a function of the atomic mass A :

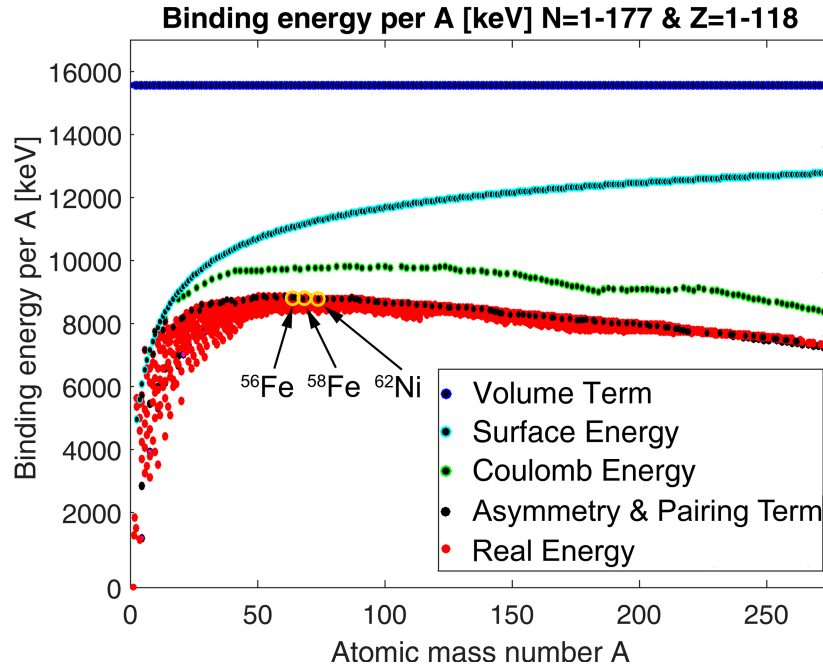


Figure 1.1: The nuclear binding energy per nucleon E_{bin}/A , is given as a function of the atomic mass A . The nuclei which are bound the strongest ^{62}Ni , ^{58}Fe and ^{56}Fe are labeled. The values of the residuals to reduce the condensation energy to the binding energy (red) are from [Wap58]. These residual energies contribute as the volume term (blue), the surface energy (cyan), the Coulomb energy (green), as well as the asymmetry term combined with the pairing term (black).

The constants of the Bethe-Weizsäcker formula Eq. (1.11) a_V , a_S , a_C , a_A and $a_\delta(A, Z)$ reflecting residual contributions, which will be described in the following. The values of those constants are empirically determined by a fit to experimental data [Wap58].

- The volume term $a_V A$ represents the condensation energy, which will be released when nucleons form a nucleus. This term has the strongest contribution to the binding energy and is proportional to the number of nucleons: $a_V = 15.58 \text{ MeV}/c^2$.
- The surface term $a_S A^{\frac{2}{3}}$ is proportional to the spherical surface of the nucleus. The reduction of the binding energy is shown by the fact that the outer nucleons are surrounded by fewer adjacent nucleons and are therefore less bound: $a_S = 18.34 \text{ MeV}/c^2$.
- The Coulomb term $a_C \frac{Z(Z-1)}{A^{\frac{1}{3}}}$, reduces the binding energy afresh. It scales with the proton number Z , due to the electrostatic Coulomb repulsion between protons: $a_C = 0.71 \text{ MeV}/c^2$.

- With the increasing difference between the proton and the neutron number the asymmetry term $a_A \frac{(N-Z)^2}{4A}$ reduces the binding energy once more⁹: $a_A = 92.86 \text{ MeV}/c^2$.
- The pairing term reflects the fact that even-even and odd-odd nuclei are more bound compared to odd-even and respectively even-odd nuclei. The later two result in a further reduction of the binding energy, in which the former contribute to the increase: $a_\delta = 11.46 \text{ MeV}/c^2$.

To illustrate how accurately the binding energy E_{bin} can be predicted by the Bethe-Weizsäcker formula, the solution has to be compared with the real measured values. If the result is plotted as in Fig. 1.2, peaks and dips are generated around the numbers, $\{Z, N\} = \{2, 8, 20, 28, 50, 82, \dots\}$. These structures indicate that the theory does not explain those effects well enough.

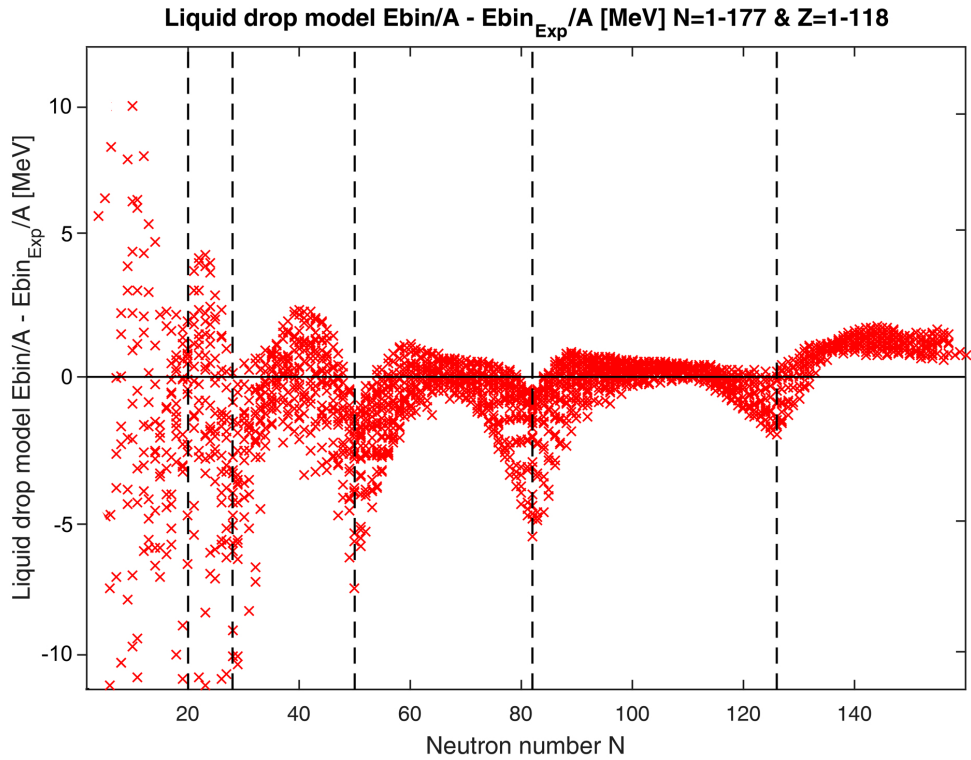


Figure 1.2: The residual binding energy per nucleon after the reduction of the predicted value of the Bethe-Weizsäcker formula, plotted as a function of the neutron number N . The peaks and dips in the plot represent the magic neutron numbers.

These effects require microscopic descriptions in order to account for the discrepancies between the model and the data. M. Goeppert-Mayer and J. H. D. Jensen

⁹Instead of the determine the constant a_A with the fit method of the empirical nucleon mass, a_A can also be determined through the Fermi-gas model and the contribution of the spin states between protons and neutrons.

proposed independently an average potential around 1950 [May48; Jen49], which was summarized in [MJ55], as a microscopic description. This description can reproduce the behavior of closed shell nuclei or often called, nuclear magic numbers. The shell structure in nuclei was determined theoretically and has thus become a firm foundation of the nuclear structure theory to this day.

1.4.2 Nuclear shell model

Apart from the macroscopic approach due to the liquid-drop model Section 1.4.1, the microscopic part was described as the pairing effect in an independent-particle model by J. H. D. Jensen and M. Goeppert-Mayer, in which it is assumed that the nucleons move inside a potential well independently from other nucleons. By suppressing the nucleon-nucleon interaction, new residual behavior were established which could be described by a mixture of a square well and an harmonic oscillator; the so called Woods-Saxon potential [WS54]. To reproduce those energy gaps of the stronger bound nucleons (magic numbers) theoretically in the shell model perspective, an ingredient named the spin-orbital interaction, described in Section 1.2, has to be added to the mean-field potential. This spin-orbit coupling, splits the states of the same orbital angular momentum, with quantum number l , into two, with total single-particle angular momenta $j = l + 1/2$ and $j = l - 1/2$. This independent-particle model¹⁰, was later enhanced mainly by two major microscopic models: The nuclear shell model [Cau05] and the Hartree-Fock-Bogoliubov approach [BHR03]. The mean-field approximation¹¹ describes a many-body complex system, which strongly interacts with each other, into a simpler, non-interacting one-body problem (quasiparticles) in an external physical potential $v(r)$, the mean-field¹².

Instead of creating a many-body Schrödinger equation, with the wave function ψ_0 :

$$H_{MF}\psi_0(r_1, r_2, \dots r_A) = E\psi_0(r_1, r_2, \dots r_A), \quad (1.12)$$

it is easier to construct the particle wave function from these one-particle states, in a sum over all one-particle energies ϵ_{α_i} with an external potential $v(r)$.

$$E = \sum_{i=1}^A \epsilon_{\alpha_i}, \quad (1.13)$$

$$h(r) = t(r) + v(r) = \frac{-\hbar^2}{2m_N} \nabla^2 + v(r). \quad (1.14)$$

The mean-field composes to $v(r)$ including the residual external potentials for the

¹⁰The independent-particle model is also called **jj**-coupling for almost all spin-orbit interactions in the nucleus, which are much stronger than those in atoms.

¹¹The content of theory in the following section was adopted from [Suh07].

¹²A field created by the surrounding A-1 nucleons.

Coulomb interaction $v_C(r)$, the nuclear spin-orbit interaction $v_{LS}(r)$ and the Woods-Saxon potential $v_{WS}(r)$. This results in a simpler description of the Hamiltonian than with the many-body solution Eq. (1.12). The kinetic energy in the Hamiltonian is represented with $t(r)$, whereas the mass m_N in the Hamiltonian is either the mass of a proton or a neutron.

To determine the optimal mean field $v(r)$ that minimizes the residual interaction between the quasiparticles, the Rayleigh-Ritz variational method, to find the minimal ground-state energy of the nucleus, either in iteration with the Hartree or the Hartree-Fock method, has to be applied. Those steps are explained explicitly in [Sub07]. After a set of eigenstates has been found, the potential can be described by the following residual potentials.

The Woods-Saxon potential $v_{WS}(r)$:

$$v_{WS}(r) = \frac{-V_0}{1 + e^{(r-R)/a}}. \quad (1.15)$$

Its usual parametrization is the nuclear radius $R = r_{0,at}A^{1/3}$, the value a as the surface diffuseness, $r_{0,at}$ as the atomic radius and V_0 as the Fermi-energy potential depth, created by the particle number density.

The Coulomb potential $v_C(r)$ can be described by an uniformly charged sphere of radius R . Due to the Coulomb repulsion this term is just related to the protons; for neutrons it is zero:

$$v_C(r) = \frac{Ze^2}{4\pi\epsilon_0} \begin{cases} \frac{3-(r/R)^2}{2R} & , r \leq R \\ \frac{1}{r} & , r > R. \end{cases} \quad (1.16)$$

Where ϵ_0 in the Coulomb potential represents the permittivity of free space.

The spin-orbit term $v_{LS}(r)$ includes:

$$v_{LS}(r) = v_{LS}^{(0)} \left(\frac{r_{0,at}}{\hbar} \right)^2 \frac{1}{r} \left[\frac{d}{dr} \frac{1}{1 + e^{(r-R)/a}} \right]. \quad (1.17)$$

The derivative in the second parentheses creates the spin-orbit generated peaks and dips in the nuclear surface region, which were shown before in Fig. 1.2. The strength term $v_{LS}^{(0)} = 0.44 \cdot V_0$, is a constant term derived from the Woods-Saxon parameters. The Hamiltonian generates the angular momentum dependence of the wave function, which contains the terms $\mathbf{L}^2$ and $\mathbf{L} \cdot \mathbf{S}$.

$$\mathbf{L}^2 |l \frac{1}{2} j m_j\rangle = l(l+1)\hbar^2 |l \frac{1}{2} j m_j\rangle, \quad (1.18)$$

$$\mathbf{L} \cdot \mathbf{S} |l \frac{1}{2} j m_j\rangle = \frac{1}{2} \left[j(j+1) - l(l+1) - \frac{3}{4}\hbar^2 \right] |l \frac{1}{2} j m_j\rangle. \quad (1.19)$$

The final mean-field Schrödinger equation composes to¹³:

$$\begin{aligned}
 h |nl\frac{1}{2}jm_j\rangle &= \frac{-\hbar^2}{2m} \left[\nabla_r^2 - \frac{l(l+1)}{r^2} \right] + v_{WS}(r) + v_C(r) \\
 &\quad + \frac{1}{2}[j(j+1) - l(l+1) - \frac{3}{4}]\hbar^2 \cdot v_{LS}(r) |nl\frac{1}{2}jm_j\rangle \\
 &\equiv h_{lj}(r) |nl\frac{1}{2}jm_j\rangle \\
 &= \epsilon_{nlj} |nl\frac{1}{2}jm_j\rangle.
 \end{aligned} \tag{1.20}$$

With this obtained knowledge it is now possible to create a schematic plot of the Hamiltonian as presented in Fig. 1.3.

The introduced nuclear shell model takes into account the nucleon-nucleon interactions, considering the interaction between valence nucleons and an inert core consisting of all other nucleons. The shell model approach to describe the proton and neutron interaction for the neutron closed-shell $N = 50$ nucleus ^{79}Cu , will be explained in more detail in Section 3.2.

For the sake of understanding, it should also be noted in the following that the deficit of the above-mentioned shell model is that it is only suitable at or close to closed shell nuclei in proton and neutron shells. In order to extend the independent-particle model, an additional possibility was announced as the *ab-initio* method of Hartree-Fock (HF), which searches for the exact Schrödinger solution by varying the nuclear mean-field to derive the single-particle states. Taking also pairing effects into account suppresses the behavior that every doubly-magic nucleus with only one nucleon more would result in a deformed nucleus. The HF model has been extended by the Hartree-Fock-Bogoliubov (HFB)¹⁴ method with the so called Wigner term as a correction factor for $N = Z$ nuclei regions. With HFB the masses over the whole nuclear chart can be predicted, with an uncertainty of about $\delta m \sim 500$ keV [Gor16], wherein with the shell model the determination of the single-particle and the spin states is provided. Specific semi-empirical mass theories like Myers-Swiatecki [MS66] or the mass formula of Duflo-Zuker [Duf95], which will be not described in this thesis, are as well very successful. For a more detailed description the reader is referred to [LPT03].

¹³The energy ϵ_{nlj} does not depend on the projection quantum number m due to the fact that all directions are equivalent for a spherically symmetric Hamiltonian. This statement can be proven by means of the Wigner-Eckart theorem.

¹⁴The formalism of BCS (Bardeen, Cooper, Schieffer), which describes the theory of superconductivity, led to the description of HFB, with the ansatz that the nucleons behave independent from each other (bosonic) and thus nucleon pairing is possible, e.g. description of superfluidity in neutron stars or ^4He .

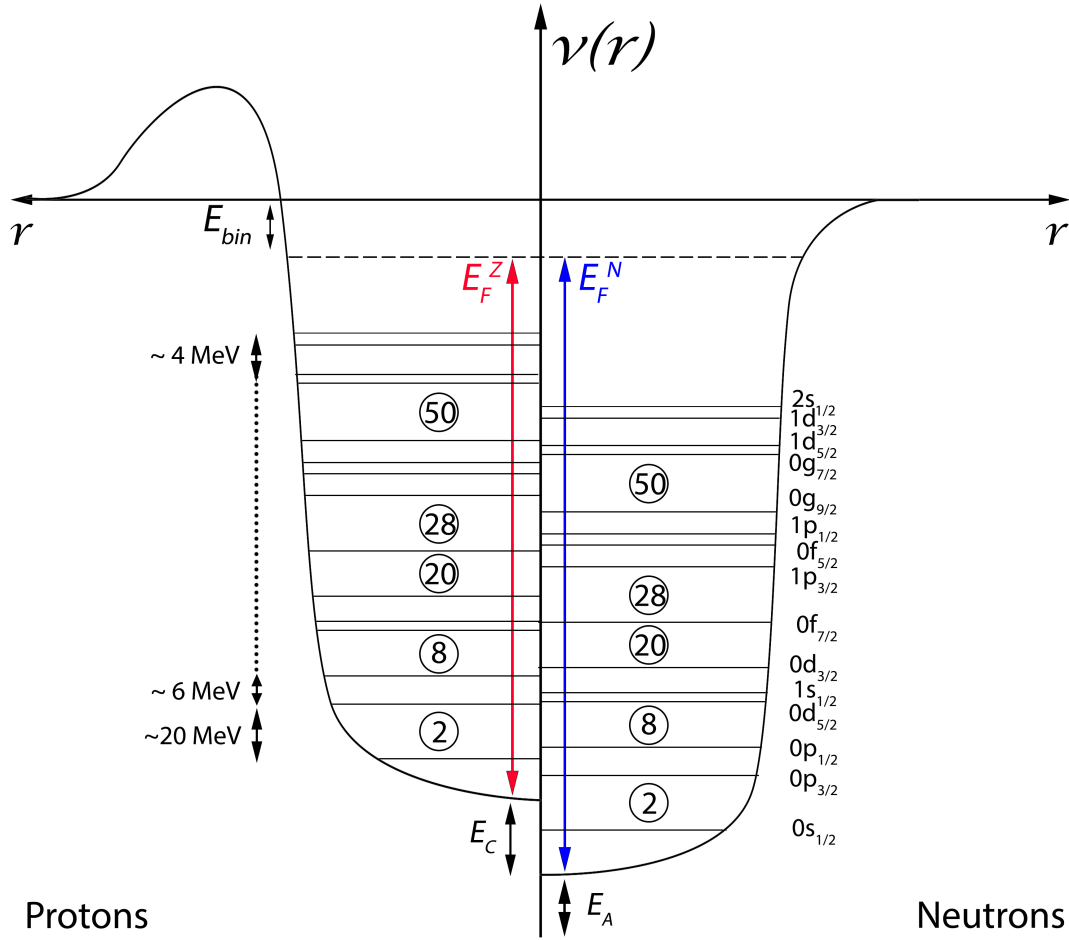


Figure 1.3: The figure represents the effective Hamiltonian $h_{lj}(r)$, from the previous described Section 1.4.2. Where the numbers surrounded by a circle are the magic numbers, illustrating the shell gap. Each line represents a sub shell, either in the proton p or the neutron n potential, labeled after the quantum state principle $|nl\frac{1}{2}jm_j\rangle$, with their respective degeneracy of $2j + 1$. Further orbitals above the magic number 50 exist and have not been listed for the sake of clarity. The split between the protons and neutrons is caused by the repulsive Coulomb potential E_C . In every Wood-Saxon potential, the difference between each sub shell is almost similar, up to $Z, N = 80$. This statement can be proven by studying mirror nuclei. The Fermi-energies in the proton E_F^Z (red), respectively, in the neutron E_F^N (blue) potentials, as well as the asymmetry term E_A are highlighted. The orbitals are not to scale.

Further access to unmeasured masses, which are close to known masses, can be achieved with extrapolations. A type of this access is a local mass formula, developed by G. T. Garvey and I. Kelson [GK66], which takes the neighboring nuclides into account to predict the mass of an unknown. In the following Section 1.5 some for this thesis relevant finite-difference mass formulas will be described.

1.5 Finite-difference mass formulas

The binding energy, which was introduced at the beginning of Section 1.3 in Eq. (1.10), contains information that can reveal the structure of a nucleus. By observing the nucleus' total binding energy, a small change, adding or reducing, about one proton or neutron has little impact and would not reveal fine details of the nuclear-structure. To approach the nuclear-structure, a numerical method for solving differential equations by approximating them with difference equations is used. This discretization methods are based on the Taylor expansion. The so called finite-difference mass formulas are modern tools in nuclear physics to visualize the fine differences in the nuclear mass-surface of a nucleus. Some examples are given bellow, e.g., the one-neutron separation energy S_n , the two-neutron separation energy S_{2n} or the empirical pairing-gap Δ_n , respectively, all following aspects apply also to protons. The introduction of the theory is preceding the explanation of the plots, which are implemented at the end of this chapter in Fig. 1.4. The following information are summarized from [Man15].

1.5.1 Separation Energies

The difference in the binding energies of neighboring nuclides reflects the energy needed to reduce a certain number of neutrons from the nucleus. The simplest approach is the one-neutron separation energy S_n .

$$S_n(Z, N) = E_{bin}(Z, N) - E_{bin}(Z, N - 1). \quad (1.21)$$

The odd-even staggering effect, seen in Fig. 1.4 a), has its origin in the enhanced binding energy of two paired neutrons. This effect is already implemented in the Bethe-Weizsäcker mass formula represented with the $\delta(A, Z)$ pairing term. If the binding energy reaches $S_n(Z, N_{drip}) = 0$, adding more neutrons to a nucleus wouldn't be possible, due to the missing binding energy, the so called neutron drip-line is reached. The sharp drop of several MeV, seen in the separation energy, highlights the effect of the magic numbers. To emphasize this effect and to suppress the odd-even staggering, dominant in the S_n , the two-neutron separation energy S_{2n} , which is able to give hints to local microscopic nuclear structure effects, is the choice.

$$S_{2n}(Z, N) = E_{bin}(Z, N) - E_{bin}(Z, N - 2). \quad (1.22)$$

The effect in the S_{2n} -plot is shown in Fig. 1.4 b). A smooth trend, lowering the binding energy along the x -axis by adding subsequently more neutrons can be observed. The effect of a shell closer, crossing a magic number, is noticeable by a sharp drop in the S_{2n} energies compared to the smooth trends beforehand.

The neutron/proton separation energies giving strong hints in the MeV regime, however to be even more sensitive to some hundreds of keV the empirical pairing

gap, a higher order finite-difference mass formula is necessary.

Binding, separation and shell-gap energies in [MeV] for the $Z=19$ (K) chain

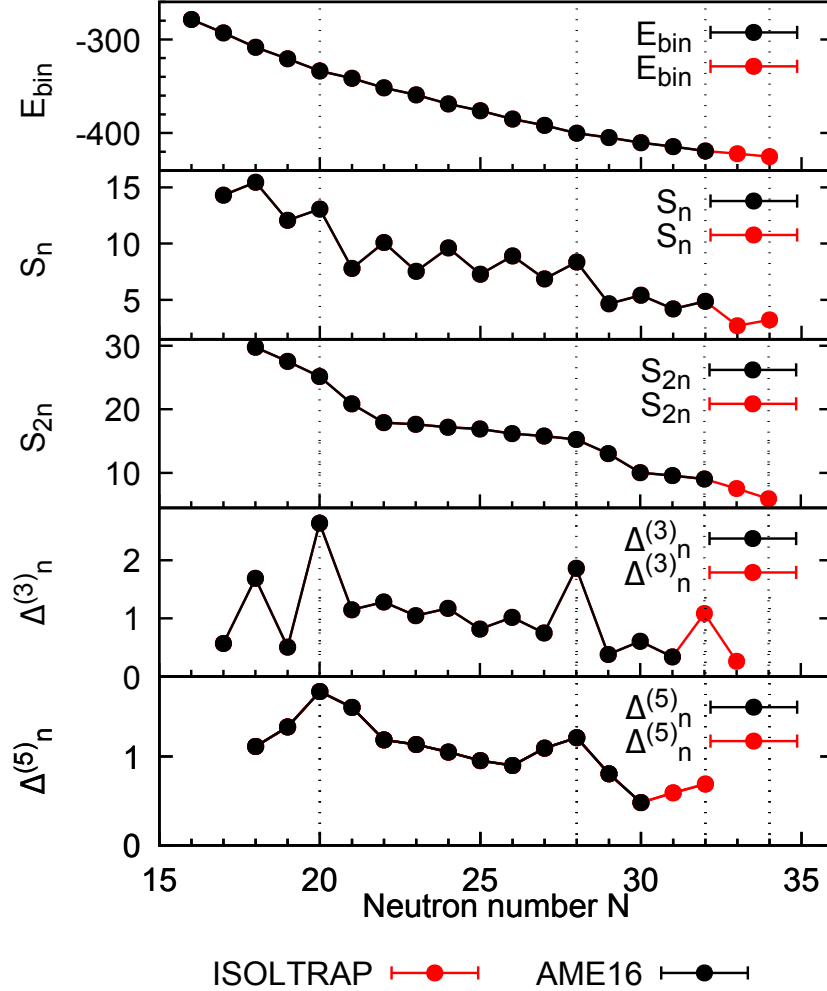


Figure 1.4: The figure represents the experimental binding energies, as well as different applied mass filters for the potassium (K) isotopes, as previously explained in the text and in reference to the presentation for calcium (Ca) in [Man15]. a) The total binding energy of a nucleus $E_{bin}(Z, N)$, compared to a certain amount of neutrons on the x -axis. The applied mass filters are: b) The one-neutron separation energy $S_n(Z, N)$. c) The two-neutron separation energy $S_{2n}(Z, N)$. d) The $\Delta_n^{(3)}(Z, N)$ pairing gap and e) the $\Delta_n^{(5)}(Z, N)$ pairing gap. The symbols in black reflecting the data from AME2016 [Wan17] and in red the recent achieved mass measurements of ISOLTRAP [Ros15].

1.5.2 The empirical pairing-gap

From the theoretical point of view the pairing gap contains more effects than just the pure pairing correlations [Ben00], e.g., the mean-field, single-particle and pairing effects. The perspective of two-different points (first order) formula, leads to the one-nucleon separation energy, which mixes every effect too strong with each other and should not be used to determine the pairing-strength. Instead the three-point difference equation and higher order formulas mainly represent the pairing effect and additionally, e.g. deformation:

$$\Delta_n^{(3)}(Z, N) = \frac{\pi_{N_0}}{2} ((E_{bin}(Z, N-1) + E_{bin}(Z, N+1) - 2E_{bin}(Z, N))). \quad (1.23)$$

Where $\pi_{N_0}=(-1)^{N_0}$ is the number parity. The odd-even staggering effect, presented in Fig. 1.4 c) in the $\Delta_n^{(3)}(Z,N)$ -plot occurs due to the stronger binding effect of even- N nuclei. The number in brackets N_0 is the point-indication parameter, how many nucleons are involved in the described interaction. Another positive aspect of the $\Delta_n^{(3)}(Z,N)$ pairing gap is that it is less biased by the Wigner effect¹⁵ [CQW15]. The four-point indicator $\Delta_n^{(4)}(Z,N)$ is antisymmetric and will not be presented here.

The best decoupling from the mean-field is achieved with the five-point gap and leads to the most robust empirical pairing [Ben00], shown in the following formula:

$$\Delta_n^{(5)}(Z, N) = \frac{\pi_{N_0}}{8} ((E_{bin}(Z, N+2) - 4E_{bin}(Z, N+1) + 6E_{bin}(Z, N) - 4E_{bin}(Z, N-1) + E_{bin}(Z, N-2))). \quad (1.24)$$

Higher orders start to smear out the staggering effects more and more, whereas the width of the peak, which represents the shell closure gets broader, due to more involved binding-energies, as shown in Fig. 1.4 d).

The strength of the finite-difference mass formulas as a tool to describe the specific contributions in the nuclear structure, can be substantiated with other physical observables, e.g., charge-radii and electromagnetic transitions, to verify the physical origin from different perspectives.

To exploit the full strength of those mass formulas in order to explore the nuclear structure, the precise knowledge of the binding energy with a small uncertainty is of great importance. The procedure and tools used at the ISOLTRAP experiment to achieve such a required accuracy, are summarized in detail in the following Chapter 2.

¹⁵The Wigner effect gives additional binding energy in case of $N = Z$ nuclides. A unique aspect of that region caused by the fact that neutrons and protons occupy the same shell-model orbitals. Thus, the large spatial overlaps between neutron and proton single-particle wave functions increase neutron-proton correlations, e.g. the neutron-proton pairing [Sat97].

Chapter 2

The ISOLTRAP experiment

Since three decades the high-precision mass spectrometer ISOLTRAP (Isotope Separator On-Line TRAP) [Muk08; Kre13; Klu13; Lun17], located at the ISOLDE [Kug00; Bor16] facility at the European Organization for Nuclear Research CERN (Geneva, Switzerland), obtained data of more than 400 exotic nuclides in the fields of mass-spectrometry, with half-lives as short as 48 ms [Man13], reaching relative mass uncertainties of about $\Delta m/m = 8 \cdot 10^{-9}$ [Kel03]. Recently ISOLTRAP extended its portfolio to include hyperfine-structure measurements [Mar13; Mar18], half-life measurements [WW16], as well as trap-assisted α , β and γ -spectroscopy of isomeric and ground states [Kow12; Sta13; AW17]. This chapter provides a brief overview of the tools used to achieve the measured masses, with the ISOLDE facility and the ISOLTRAP experiment being discussed in detail.

2.1 The ISOLDE facility, a radioactive ion beam facility

The ISOLDE facility at CERN [Kug00], is dedicated to the production of short-lived nuclides to study a broad range of physics, such as nuclear-, atomic-, astro- and solid state physics, as well as applications in life science. To elucidate the ISOLDE facility an overview is given in Fig. 2.1.

The most common reactions in which radionuclides are produced are spallation, fragmentation or fission. These reactions are induced by a 1.4 GeV bunched proton beam. Each bunch consists of about $3.2 \cdot 10^{13}$ protons which are separated in a 1.2 s pulsed structure. The bunched proton beam is delivered from the Proton Synchrotron Booster (PSB) with an intensity of up to 2.1 μ A. These proton bunches impinge on a thick target, usually UC_x, generating a cocktail of different nuclides along the nuclide chart with lower masses as the target material. To suppress neutron-deficient contaminants a tungsten cylinder is impinged by the proton beam instead, which acts as a neutron converter. The neutron-rich isotopes are then produced by neutron-induced fission [Got14]. The ISOLDE facility hosts two

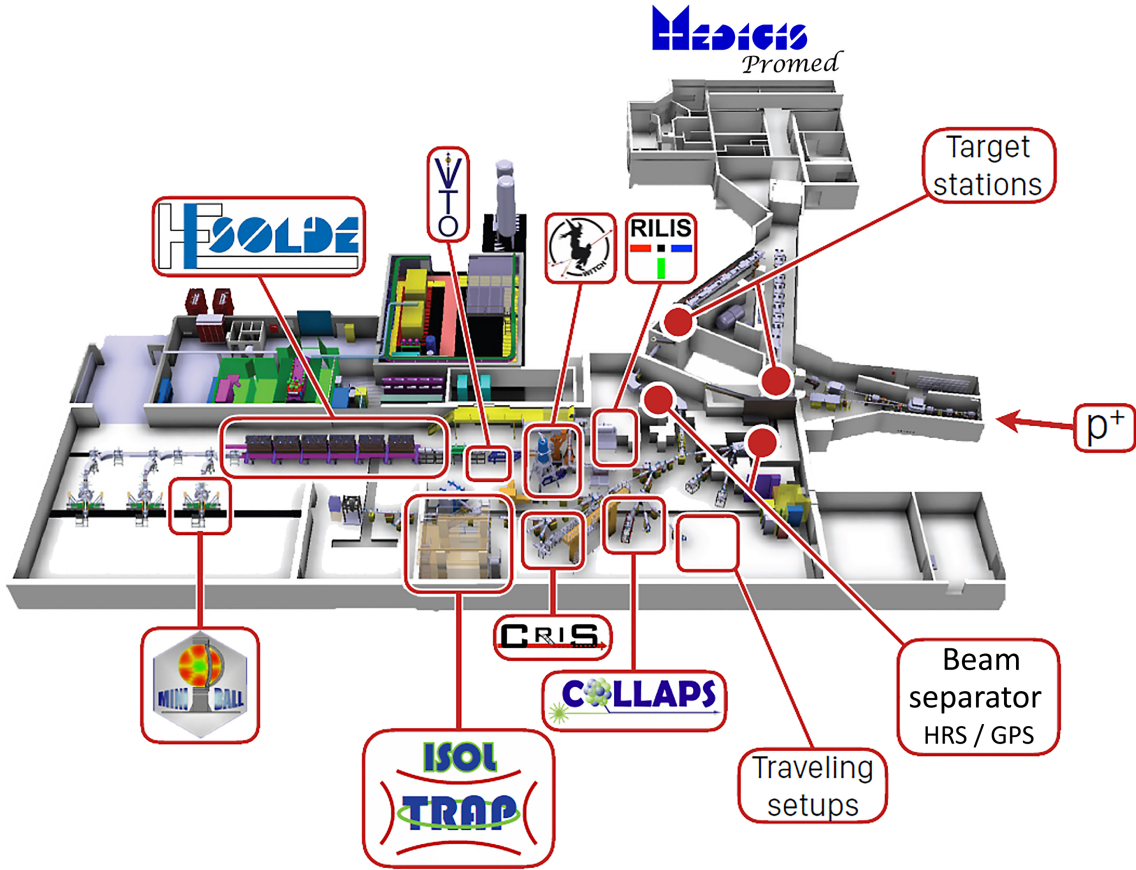


Figure 2.1: The ISOLDE facility with its experiments [Kug00]. The protons are delivered with an energy of 1.4 GeV from the Proton Synchrotron Booster (PSB). To produce the radioactive nuclides the protons impinge on a thick target, usually UC_x , either in the High Resolution Separator (HRS) or the General Purpose Separator (GPS) target section, the mass filters with a mass resolving power of $m/\Delta m = 5000$ and $m/\Delta m = 1000$, respectively. After the ions are ionized and mass separated, they are guided with electro-static potentials through the beam line into the setups, e.g. the ISOLTRAP experiment. Picture credits [Kar17].

target areas named after the separators, namely General-Purpose Separator (GPS) and High-Resolution Separator (HRS). By use of different types of targets as well as a broad spectrum of extraction and ionization techniques, a large variety of nuclides from $Z = 2 - 92$ with half-lives down to ms are feasible at ISOLDE. Fig. 2.2 represents, in the form of the periodic table of Mendeleev [Men69], the elements which are available at the ISOLDE facility.

Group	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
	1A	2A	3B	4B	5B	6B	7B	8B			1B	2B	3A	4A	5A	6A	7A	8A	
Period																			
1	1 H																	2 He	
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne	
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
6	55 Cs	56 Ba	*	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	**	103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg							
* Lanthanides			*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
** Actinides			**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		

Figure 2.2: A periodic table with the elements available as radioactive beams at ISOLDE. The colors indicate the corresponding ion source. The elements with a white background are not yet produced. (database of ISOLDE target group [Iso])

After the atoms are ionized, the isotopes are accelerated to 30-60 keV and guided through the separator magnets. Depending on which target area they are produced, the separator is either the GPS with a mean mass-resolving power of $m/\Delta m = 1000$ or the HRS with the ability to resolve different masses with an even higher mean mass resolving-power of $m/\Delta m = 5000$ ¹. The physics behind the separation of charged particles passing a static magnetic field is the Lorentz force. After the isobaric separation phase, the ion beam is transported to various experiments situated at ISOLDE via a beam-line equipped with electrostatic ion-optical elements. At the end of the central beam-line the high-resolution mass-spectrometer ISOLTRAP is located, which will be described in the following.

2.2 The ISOLTRAP experiment

At the end of the central beam-line, the mass-selected nuclides, are delivered as a 30-60 keV continuous ion-beam to the ISOLTRAP setup, which consists currently of four ion traps. In the first trap of ISOLTRAP the ions are stopped, cooled by interactions with helium buffer gas and bunched in a linear Radio-Frequency Quadrupole (RFQ) [Her01]. At this stage a high amount of unwanted nuclides might be still present in the bunch. In order to prepare a bunch with predominantly nuclides of

¹The mass resolving-power is a measure of how good neighboring isobars (masses with same number of nucleons $A = N + Z$) can be separated.

interest, from a possible mixture of nuclides, a multi-reflection time-of-flight mass separator (MR-ToF MS) [Wol12; Wol13b] with a typical mass resolving-power of $\mathcal{R}_{MR-ToF} = m/\Delta m = 10^5$ is used [WW15]. Thereafter, the ion bunch is transferred to the cylindrical Penning trap [RH97] for cooling, re-centering and additional isobaric cleaning by mass-selective resonant buffer-gas cooling [Sav91]. The further purified and cooled bunch of ions is then injected into a hyperbolic precision Penning trap. At this stage in the fourth trap, the ions' mass will be measured with high accuracy.

The schematic overview of ISOLTRAP is shown in Fig. 2.3, followed by a detailed description of the apparatus.

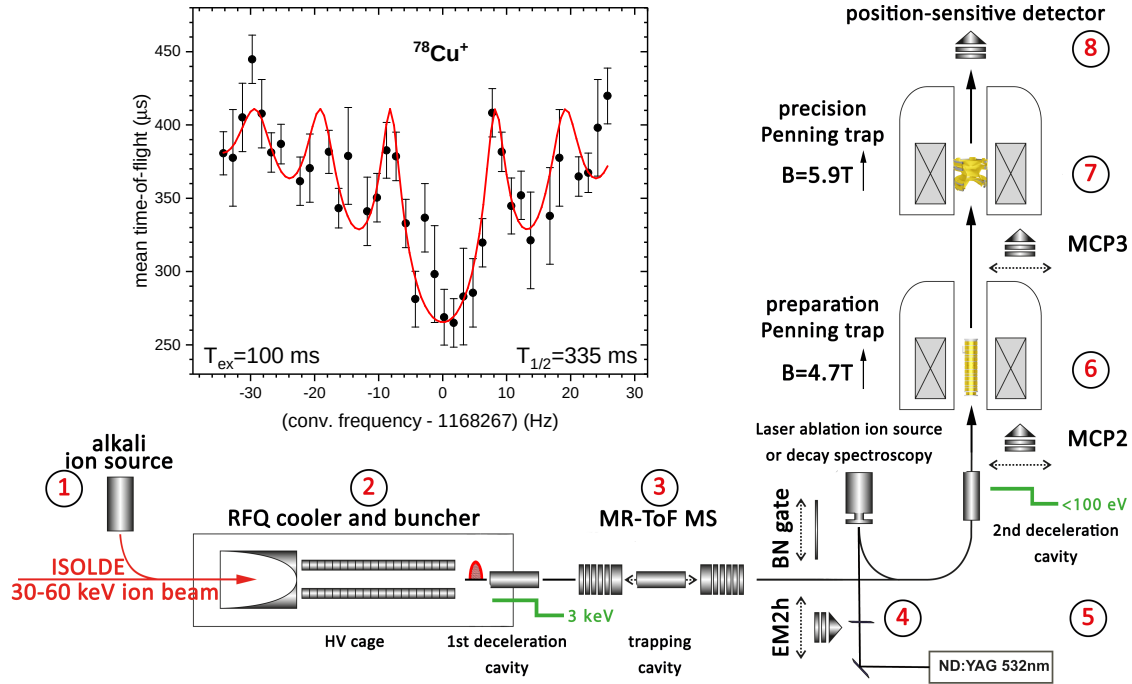


Figure 2.3: The schematic representation of ISOLTRAP can be divided into the horizontal beam line No. (1)-(5) and the vertical beam line No. (6)-(8).

The ions arriving as a 30-60 keV continuous ion-beam from ISOLDE or the offline alkaline ion-source (1). The arriving ions are accumulated, cooled and bunched in the RFQ (2). The bunched ion-beam passes the first pulsed drift tube to be de-accelerated and injected afterwards into the MR-ToF mass separator (3). After the first mass separation, the ions mass can be determined by the electron multiplier detector (EMP2h) or implanted into a decay chamber (4). Alternatively, a carbon-cluster source can be ionized by a Nd:YAG-laser (5), e.g. for systematic studies. Thereafter, the ion bunch is transferred to the cylindrical Penning trap (6) for cooling, re-centering and additional isobaric cleaning by mass-selective resonant buffer-gas cooling. Prior that stage the beam is further decelerated in the second pulsed drift tube, to achieve an ion energy below < 100 eV. The cleaned ion bunch is then injected into the hyperbolic precision Penning trap (7). Here, the high-precision

mass measurement with either the time-of-flight ion-cyclotron resonance (ToF-ICR) or the phase-imaging ion-cyclotron-resonance (PI-ICR) technique is achieved by use of the position-sensitive multi-channel plate detector (MCP) on top of ISOLTRAP (8). The inset shows a typical ion-cyclotron resonance, taken with a $T_{ex} = 100$ ms cyclotron excitation time.

2.2.1 Radio-Frequency Quadrupole

As previously mentioned, the up to 60 keV ion beam from ISOLDE or from - ISOLTRAP's alkaline reference ion source, reaches ISOLTRAP as a quasi-continuous beam. To be able to perform a measurement with Penning traps, well defined bunches have to be formed. The thermal energy spread of the formed ion cloud is reached by cooling the ions' motion and with it the reduction of the beam emittance. The cooling is based on the following principle: The continuous beam is electrostatically retarded to an energy of a few eV, followed by the injection into the segmented linear Radio-Frequency Quadrupole (RFQ) trap, which consists of four rods of the same kind. To reduce the energy of the injected ions, the RFQ trap is filled with helium, which acts as a buffer gas at a pressure of roughly $\sim 10^{-3}$ mbar. This results in elastic scattering and interaction of the dipole moments between the ions and the slightly polarized helium atoms [Ell78]². With the exact cooling time, the ions will lose their kinetic energy down to the thermal equilibrium of the buffer-gas atoms. In case of ISOLTRAP down to room temperature $T = 293$ K. In order to maintain control over the ion cloud, a potential well is created with different DC (direct current) voltages in which the ions can settle. These voltages are applied along the axis to each segment, defining a guiding gradient. If these DC voltages are imposed by a radio-frequency (rf), a three-dimensional pseudo-potential well will be formed, providing a radial confinement³. This relation is demonstrated as an overview in Fig. 2.4. For further details about the RFQ cooler and buncher, the reader is referred to [Her01].

As a result of the minor influence of the elastic scattering in the cooling process for thermal energies E_{therm} and the major effect of the dipole interaction, the buffer-gas cooling can be described by the viscous-drag model [Sav91], as in the following relation. Introducing the damping force $\mathbf{F}_\delta$, which can approximately written as:

$$\mathbf{F}_\delta = -\delta \cdot m \cdot \mathbf{v}, \quad (2.1)$$

wherein $m\mathbf{v}$ is the momentum of the ion and δ is the damping parameter, which is described by the physical parameters:

$$\delta = \frac{q}{m} \frac{1}{K_0} \frac{p/p_N}{T/T_N} \quad (2.2)$$

²The neutral, closed-shell helium atom is slightly polarized by the ion Coulomb force which contributes to a dipole type interaction.

³The radial confinement in a quadrupolar field can be described in terms of the Mathieu equations [Pau90], which also defines the stability criteria of the motion.

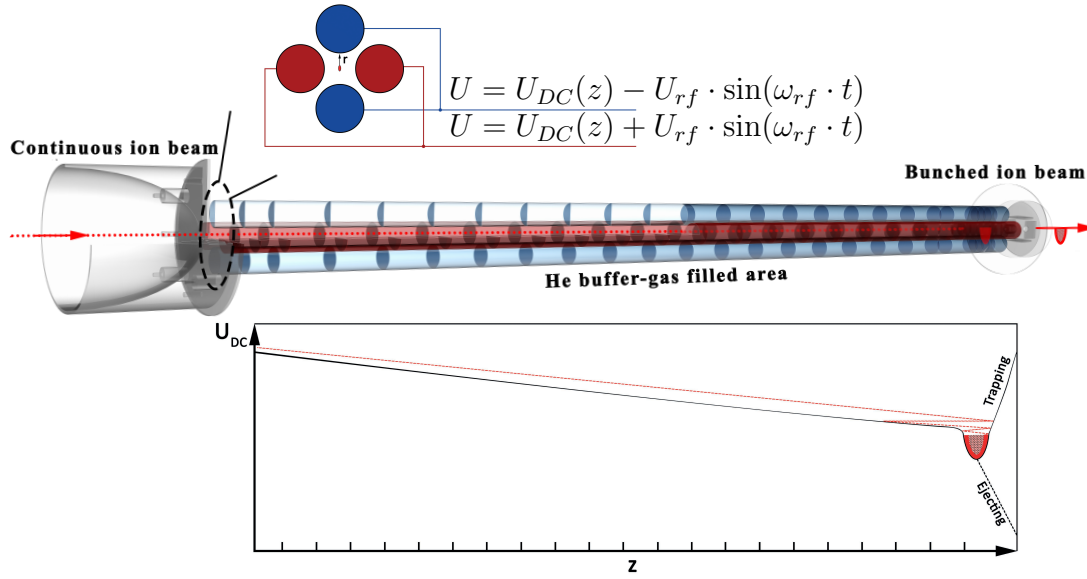


Figure 2.4: A side-view of the segmented radio-frequency quadrupole trap with the length z , acting as a cooler and buncher at the ISOLTRAP experiment. The trap is filled with helium buffer-gas to reduce the incoming ion beam's energy, to a thermal equilibrium at $T = 293$ K, respectively, $E_{therm} = 0.025$ eV. The illustrative 2D-trapping and ejection potentials of the four rods are sketched beneath the trap. In order to keep the ions centered, a radio-frequency U_{rf} is applied, which is superimposed by the U_{DC} potential, creating a 3d pseudo-potential.

The damping factor takes the ion mobility K_0 , the charge-to-mass ratio of the ion $\frac{q}{m}$, the pressure p as well as the temperature of the buffer gas, as ratio to the normal pressure $p_N = 1013.25$ mbar and the normal temperature $T_N = 273.16$ K, into account. The ion mobility with kinetic energies > 1 eV decreases [LM99], therefore, at ISOLTRAP it is sufficient to take the constant low-energy values as an upper limit for the ion mobility. Within a few 10-100 ms [Her01], the ion bunch is in thermal equilibrium with the buffer gas in the potential minimum at the end of the RFQ. After the accumulation and cooling time, the ions would fly with the high potential of the platform of $U_{HV} = 30-60$ kV to the subsequent traps. To avoid this effect the RFQ is followed by a pulsed cavity tube, which can be switched to a certain voltage. Studies have shown that an end-point energy of $E_{end} = 3.2$ keV matches the capture acceptance to inject into the MR-ToF MS [Wol11], which follows the RFQ. Therefore, the ions are ejected from the RFQ by lowering the end electrode potential into the cavity, which is at a $U_{cavity} = U_{HV} - 3.2$ kV lower potential than the RFQ platform. The ions travel as an almost Gaussian shaped bunch, with a width of only a few tens of nanoseconds, into the cavity. This cavity is an electrostatic field-free region, therefore, no field gradient is generated, which could disturb the ions trajectory. After the ion bunch reaches the center of the cavity, the potential

is switched within a few tenth of nanoseconds to ground-potential. The cavity acts as a potential lift, where the resulting kinetic end-point energy is calculated as $E_{end} = q \cdot (U_{HV} - U_{cavity})$.

The mechanism of cooling an ion in a buffer gas filled Penning trap environment, is described in Section 2.2.5. Other cooling techniques, like resistive, evaporative [KD96] and electron cooling [Bud67], are described as a summary in [Ita95; Bla06]. These cooling techniques are not part of the ISOLTRAP setup and are here just mentioned for the sake of completeness.

2.2.2 Multi-Reflection Time-of-Flight Mass Separator

The consecutive step for the cooled and bunched ions, is the injection into the multi-reflection time-of-flight mass separator (MR-ToF MS). This separator is a 80 cm apparatus, which was installed at ISOLTRAP in 2010 by R. Wolf [Wol12; Wol13a]. The specialty of ISOLTRAP's MR-ToF MS, is the pulsed drift cavity, with which the energy of the ions is adjusted to the beam-line potential or the trapping potential of the device. The first milestone of this technique was set in 1990, by H. Wollnik, who suggested to use the same ion path multiple times to enhance the time-of-flight mass separation (ToF-MS) technique [WP90]⁴. An overview of the apparatus and a part of the post beam-processing is given in Fig. 2.5.

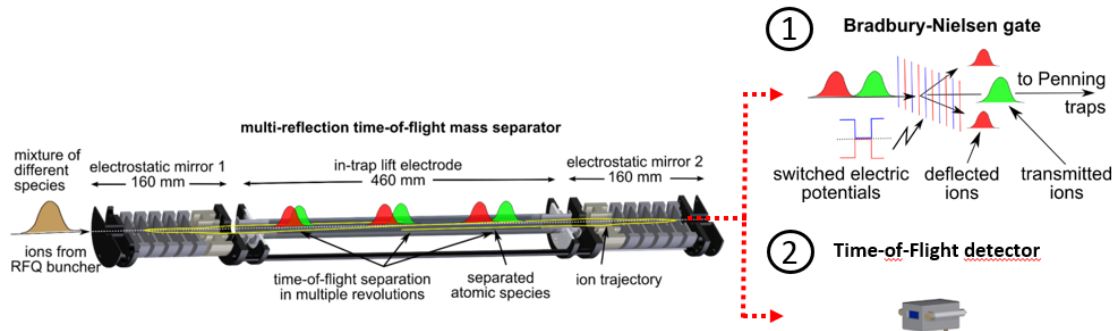


Figure 2.5: Ions are injected as an isobaric mixed bunched beam into the Multi-Reflection Time-of-Flight Mass-Separator (MR-ToF MS). The ions passing back and forth and due to their slight mass difference they might be separated. On the right-hand side two possible ejection scenarios are illustrated: 1) The Bradbury-Nielsen beam Gate (BNG) on-axis with the ejected beam, used as a beam gate for the ion-of-interest (green) and a deflector for the ions-of-non-interest (red). 2) In case of the electron-multiplier detector on-axis, the resolved beam can be analyzed and measured by the time-of-flight mass measurement technique. See text for detailed information. Drawing taken from [Wol13b].

The MR-ToF MS consists of two opposed mirror-electrode structures, with a 460 mm long central-cavity electrode, acting as an in-trap potential lift. The length of

⁴The mass separation with electrostatic potentials was achieved already before, only technical restrictions and a mass resolving power of $m/\Delta m = 20$ in 1960 by W. Tretner [Tre60] lessened the success.

one mirror set is 160 mm and consists of six mirrors. The two closer to the center cavity are supplied with a negative potential, while the other following four are defining the positive repelling mirror potential. The central cavity potential is in the trapping phase on ground potential. Only during the injection and ejection phase a pulse lifts the potential from the trapping potential to the injection or ejection potential of roughly 1 kV, by this means the kinetic energy of the ion beam matches ISOLTRAP's beam-line potential. With the higher kinetic energy of the whole ion bunch, the ions are able to overcome the mirror potentials of the MR-ToF MS, within the injection or ejection phase. With this important step only one voltage has to be switched, which leads to higher overall simplicity and voltage stability. The subsequent step for the bunched ion-beam prior the ejection is the isobaric separation in the MR-ToF MS. After multiple revolutions between the two electrostatic mirror sets of the MR-ToF MS, the separated beam can either be detected using an electron multiplier or transported further to the Penning traps.

The MR-ToF MS can be used as a beam purifier by separating the different ion species or as a mass measurement device. For the latter application, the mean time of flight between the ISOLTRAP RFQ-buncher and the detector is given by $t = \alpha_{ToF} \sqrt{(m/q)} + \beta_{ToF}$, where α_{ToF} and β_{ToF} are calibration constants. In most of the cases at ISOLTRAP, q equals the elementary charge. The obtained time-of-flight spectrum can be thus calibrated using ions of known mass to determine the mass of the ion of interest. To calibrate the MR-ToF MS, the offline alkaline source with $^{39,41}\text{K}$, $^{85,87}\text{Rb}$ and ^{133}Cs can be used. The final formula linking the mass of the ion of interest m_{IOI} to the masses of the reference ions m_{ref1} , m_{ref2} is [Wie13]:

$$m_{IOI} = (C_{ToF} \cdot (\sqrt{m_{ref1}} - \sqrt{m_{ref2}}) + 0.5 \cdot (\sqrt{m_{ref1}} + \sqrt{m_{ref2}}))^2, \quad (2.3)$$

where $C_{ToF} = (2t_{IOI} - t_{ref1} - t_{ref2}) / [2(t_{ref1} - t_{ref2})]$ is the experimental time-of-flight ratio. The mean total flight time of the ion of interest is t_{IOI} , where $t_{ref1,2}$ represent the flight times of the reference masses, respectively. With only two reference ions, this approach can be used to determine the mass with an uncertainty of approximately $\sim 10^{-6}$ [Wie13]. The reference ions have to be therefore either in the actual spectrum of the mass to be measured (online reference) Fig. 2.5, e.g. $^{65}\text{Cu}^{12}\text{CH}_2$ copper-methylene and ^{79}Ga or in a sample of an online and offline-source reference measurement (offline reference). In order to be accurate, the reference offline mass has to be measured before or after the main measurement, even if the main determining mass is the online mass within the spectrum of the mass to be measured. In addition, both masses must be at the same number of revolutions until a mathematical correction approach has been established. If measurements are performed in close proximity to each other, it can be ensured that drastic temperature drifts and strong voltage drifts of the mirrors and the cavity are less likely. Taking the above mentioned steps into account minimizes the error of the mass to be measured. Studies which will be published elsewhere have shown that within the achievable systematic uncertainties a reference in the online spectrum, close to the ion of interest to be determined, is sufficient to measure the mass by MR-ToF MS. The strength of the MR-ToF MS is to resolve close lying masses within a short time

range of about several milliseconds. Thus, ISOLTRAP's MR-ToF MS is able to approach a mass resolving power of the order $\mathfrak{R}_{MR-ToF} = 3 \cdot 10^5$, which is determined by [Wol13b; Wie17]:

$$\begin{aligned} \mathfrak{R}_{MR-ToF} &= \frac{m}{\Delta m} = \frac{t}{2\Delta t} \\ &\cong \frac{1}{2} \frac{t_{un} + n_r T_r}{\sqrt{\Delta t_{th}^2 + \left(\Delta t_E - n_r T_r \left(\frac{\partial \delta_{T_r}}{\partial \delta_E} \right) \Delta \delta_E \right)^2 + (\Delta t_A + n_r \Delta T_A)^2}}. \end{aligned} \quad (2.4)$$

Here, t_{un} is the unperturbed direct flight time between the RFQ and the electron-multiplier detector. T_r determines the revolution period within the MR-ToF MS, where the number of these is given by n_r . The uncertainties which are limiting the resolving power define the total time-of-flight spread Δt . These are, among others, the thermal time spread Δt_{th} , resulting from the trapped ions in the RFQ, which sets the lower uncertainty limit. The time spread resulting from the kinetic energy distribution of the ion bunch is described by Δt_E , where the relative deviation of this flight time per revolution is defined as $\partial \delta_{T_r} / \partial \delta_E$ combined with the relative kinetic energy spread $\Delta \delta_E$. The last part of the time-of-flight spread describes higher order divergences, outside Δt_A , respectively, inside ΔT_A the MR-ToF MS. Further comparison of the mass resolving power between different techniques used at ISOLTRAP, are described in Fig. 4.5.

The obtained mass resolved bunches with the MR-ToF MS can be treated at ISOLTRAP in two different post-procedures. In one procedure the ions can be injected into the Penning traps where further preparation or even higher accurate mass determinations can be achieved, as illustrated in Fig. 2.5 (1). In the other mode the ion events can be recorded by an electron multiplier detector on-axis with the ejected and separated ion bunch, as shown in Fig. 2.5 (2). Two distinct mechanisms are foreseen to receive a contaminant-free ion-selective ejection into the next trap or onto the electron multiplier. The old Bradbury-Nielsen beam gate method on the one hand and the novel double in-trap lift method on the other hand. Those two techniques are briefly described in the following.

- **Bradbury-Nielsen beam gate**

The Bradbury-Nielsen beam gate [BN36], highlighted with No.1 in Fig. 2.5, is a copper wire-grid. The grid can be used as an ion gate, which either has a floated potential (~ 0 V), allowing all ion species to pass through or with a repelling potential (± 250 V) to block all ions, respectively. To determine the ion species selectively, the ToF of all ions in the MR-ToF spectrum must be measured beforehand with the electron multiplier. The time information obtained is then to be used for a time window that either deflects or transmits the ions of interest after each MR-ToF ejection.

• Double in-trap lift

With the novel double in-trap lift technique, the Bradbury-Nielsen beam gate is no longer needed. The ion selection occurs by switching the central cavity. The switching time can be set that accurate, if the ion of interest is at least 250 ns far apart from contaminants. To select the different ion species, the cavity potential is only lifted at a certain moment to the ejection potential of the MR-ToF MS. The ions which are in the central cavity are able to overcome the mirror potential. The in-trap lift timing pulse window can be arranged that only ions of interest are in the center cavity, while the contaminants remain trapped in the mirror potentials [Wie17].

Since the MR-ToF MS was implemented at ISOLTRAP it is always used as a mass separator for contaminants; a beam purifier. With the two mass selective ejection techniques the Bradbury-Nielsen beam gate and the double in-trap lift technique, a preparation for the Penning trap is feasible and even more reliable. If the yield or the half-life does not allow further preparation or a more precise measurement with the Penning traps, the mass measurement with the MR-ToF MS and the electron multiplier is the first choice.

In the recent past the MR-ToF MS was used for several milestones at ISOLTRAP. Target yield checks [Got14] and contaminant checks for life science [Mue16a; Mue16b]. In combination with the RFQ trap, half-life studies [WW16], as well as purification assisted α -, β - and γ -spectroscopy after the MR-ToF MS were achieved [Sta13; AW17].

2.2.3 Introduction of Penning traps

Penning traps as state-of-the-art mass spectrometers achieved their technical break through by H. G. Dehmelt⁵ and W. Paul⁵ [Deh90; Pau90] in the mid-1970s. To these successes prior achievements of F. M. Penning in 1936 and J. R. Pierce in 1949 have played an important role. F. M. Penning increased the sensitivity of ionization vacuum gauges by use of a homogeneous magnetic field, which was arranged perpendicular to the moment of the charged particles [Pen36]. J. R. Pierce used this principle to built the first Penning trap, via superimposing the homogeneous magnetic field with an electrostatic trapping potential [Pie49]. To determine a mass really accurate by a Penning trap the knowledge of the ions' eigenfrequencies is necessary. This is advantageous, as the frequency is one of the quantities which can be determined most precisely [Sch17].

Since the Penning trap was established, the mass spectrometry was revolutionized in ever higher accuracy. In the following section, the Penning trap will be discussed in detail. The sections are separated in the ideal Section 2.2.3 and the real Penning traps Section 2.2.3, taking into account the external influences, as well as the different detection and cooling techniques relevant to the ISOLTRAP experiment.

⁵1989 Nobel Prize in physics for the development of the ion trap technique.

Ideal Penning traps

Based on the ISOLTRAP's Penning traps, the functional principles of ideal Penning traps will be introduced and later on extended by the real Penning trap.

The ISOLTRAP experiment consists of two types of Penning traps, the cylindrical preparation Penning trap, which acts as a mass selective buffer-gas cooler trap and a hyperbolic Penning trap for high-precision measurement purposes. Both traps, each rested in a strong homogeneous magnetic field, are illustrated in Fig. 2.6 a,b). The corresponding three decoupled eigenmotions of a captured ion in a Penning trap resulting from the superposition of the weak electrostatic and the strong homogeneous magnetic field are illustrated in Fig. 2.6 c).

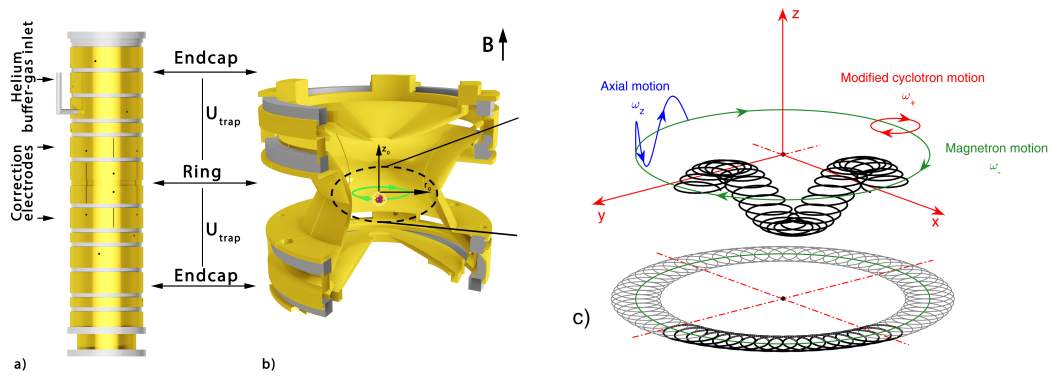


Figure 2.6: On the left-hand side a rendered picture of the cylindrical Penning trap a) is shown, followed by the hyperbolic shaped precision Penning trap b). U_{trap} defines the trapping potential between the end caps and the ring electrodes, which acts as the axial confinement. The radial confinement of the ions is achieved by the superimposed homogeneous magnetic field pointing into the z-direction. On the right-hand side a plot to exemplify the three decoupled eigenmotions c), as a result of the superposition of both fields, is given. The slow magnetron motion, the axial motion and the fast reduced cyclotron motion. A superposition of all three eigenmotions is illustrated as the black alternating line shape, where the xy-plane represents the epicyclic projection of the motions, neglecting the axial motion. In these pictures the traps are not to scale.

The strong homogeneous magnetic field aligned to the z-axis $\mathbf{B} = B\hat{e}_z$ confines the ions with the Lorentz force within the radial plane. This field is superimposed by an electrostatic quadrupole field generated by the electrode configuration of the ring and the end caps, as shown in Fig. 2.6. The resulting three dimensional electrostatic field is confining the axial degree of freedom⁶. The electrostatic trapping potential well depth of both traps at ISOLTRAP is defined by the voltage between the ring and end cap electrodes with $U_{trap} \approx -10$ V. While the magnetic field strength for the preparation Penning trap is $\mathbf{B}_{PPT} = 4.7$ T, the one for the high-precision Penning trap achieves $\mathbf{B}_{HPT} = 5.9$ T. To reach such high static magnetic fields at ISOLTRAP superconducting magnets are used. Each trap is based in a room-temperature vacuum

⁶For the electrical potential, in both trap configurations, it is not possible to confine all directions as a consequence of the Laplace equation [BG86].

tube, surrounded by charged solenoid wire-coil rested in a liquid-helium cryostat, to preserve the superconducting state of the magnet. An example of high-precision measurements, in which also the trap is cryogenically cooled is explained in [Ulm13]. A trapped particle with the charge q experiences in an ideal Penning trap the total force:

$$\begin{aligned}\mathbf{F}_{tot} &= \mathbf{F}_{lor} + \mathbf{F}_{el} \\ &= q(\mathbf{v} \times \mathbf{B}) + q\mathbf{E}(\mathbf{r}),\end{aligned}\tag{2.5}$$

where $\mathbf{F}_{lor}$ is the Lorentz force and $\mathbf{F}_{el}$ the electrostatic force, defined as:

$$\mathbf{F}_{el} = -q\nabla U(\mathbf{r}).\tag{2.6}$$

The trap potential $\Phi(\mathbf{r})$ is given by

$$\Phi(\mathbf{r}) = \Phi(r, z) = \frac{U_{trap}}{2d^2} \left(z^2 - \frac{r^2}{2} \right).\tag{2.7}$$

The radial distance of the stored particle from the trap center is defined by r , whereas the distance in the axial direction is z , leading to the characteristic trap dimension d :

$$d^2 = \frac{1}{2} \left(z_0^2 + \frac{1}{2}r_0^2 \right).\tag{2.8}$$

The shortest distance between the electrodes and the center of the trap in the radial plane is described by $r_0 = 13$ mm, as well as the axial direction as $z_0 = 11.18$ mm in case of the precision Penning trap [Muk08]. Trapped charged particles within a magnetic field $\mathbf{B}$ and a mass m can be described by three decoupled characteristic eigenmodes, shown in Fig. 2.6 c), which can be solved analytically by classical [BG86; Kre92] or quantum mechanical solutions [Sok67].

The cyclotron frequency of a particle in a magnetic field is defined as:

$$\omega_c = \frac{q}{m}\mathbf{B}.\tag{2.9}$$

The cyclotron frequency Eq. (2.9) can be included into Eq. (2.6), combined with the fact that the magnetic field is parallel to the axis of the trap, $\mathbf{B} = B\hat{\mathbf{e}}_z$ in Eq. (2.5) can be exchanged. The resulting equations are the: two-dimensional radial motion,

$$\ddot{x} = \dot{y}\omega_c + \frac{1}{2} \left(\frac{qU_{trap}}{md^2} \right) x,\tag{2.10}$$

$$\ddot{y} = -\dot{x}\omega_c + \frac{1}{2} \left(\frac{qU_{trap}}{md^2} \right) y,\tag{2.11}$$

and the one-dimensional axial motion,

$$\ddot{z} = -\frac{1}{2} \left(\frac{qU_{trap}}{md^2} \right) z.\tag{2.12}$$

Therefore, because in an ideal Penning trap, radial and axial motions are decoupled, the motions can be described by means of harmonic oscillators. By consequence, the axial motion can be expressed by the Eq. (2.12) to the squared oscillation frequency:

$$\omega_z = \sqrt{\frac{qU_{trap}}{md^2}}. \quad (2.13)$$

Using the ansatz $u = x + iy$ with $u = e^{-i\omega t}$, the radial motion coefficients are:

$$\omega^2 - \omega_c \cdot \omega + \frac{1}{2}\omega_c^2 = 0, \quad (2.14)$$

resulting to the radial eigenfrequencies:

$$\omega_{\pm} = \frac{1}{2} \left(\omega_c \pm \sqrt{\omega_c^2 - 2\omega_z^2} \right). \quad (2.15)$$

Where ω_z is the axial motion, ω_- the magnetron motion and ω_+ the reduced cyclotron motion.

With Eq. (2.13) and Eq. (2.15) the following relations can be deduced:

$$2\omega_+ \cdot \omega_- = \omega_z^2, \quad (2.16a)$$

$$\omega_+ + \omega_- = \omega_c, \quad (2.16b)$$

$$\omega_+^2 + \omega_-^2 + \omega_z^2 = \omega_c^2, \quad (2.16c)$$

Eq. (2.16b) is just true for ideal Penning traps, while Eq. (2.16c) very accurately satisfied also in the real Penning trap case [BG86].

The trapping conditions $\omega_c > 2\omega_z^2$ and $qU_{trap} > 0$ in order to store the ions must be accomplished. The two radial frequencies Eq. (2.15), can be written as a function of the trap parameters, including the cyclotron frequency coefficients:

$$\omega_{\pm} = \frac{1}{2}\omega_c \left(1 \pm \sqrt{1 - 2\frac{\omega_z^2}{\omega_c^2}} \right), \quad (2.17)$$

$$= \frac{qB}{m} \left(1 \pm \sqrt{1 - 2\frac{m}{q} \frac{U_{trap}}{d^2 B^2}} \right). \quad (2.18)$$

An overview of the typical eigenfrequencies at ISOLTRAP are represented in Table 2.1.

From the consequence that the magnetic field is much stronger than the electric quadrupole potential, the following conclusion can be drawn, $\omega_+ \approx \omega_c$, as reflected in the values of Table 2.1. Thus, the frequency hierarchy of a Penning trap can be determined to: $\omega_c > \omega_+ \gg \omega_z \gg \omega_-$.

With this, the square root in Eq. (2.18) can be approximated by the first-order Taylor expansion ($\sqrt{1 \pm x} \approx 1 \pm \frac{x}{2}$ if $x \ll 1$) to:

$$\omega_+ \approx \frac{qB}{m} \left(1 - \frac{m}{q} \frac{U_{trap}}{2d^2 B^2} \right), \quad (2.19)$$

$$\omega_- \approx \frac{U_{trap}}{2d^2 B^2}. \quad (2.20)$$

Table 2.1: Typical ion motion frequencies achieved with the ISOLTRAP's precision Penning trap ($B = 5.9$ T). Examples for the singly charged ions $^{21}\text{Ne}^+$, $^{85}\text{Rb}^+$, $^{133}\text{Cs}^+$ and the singly charged molecule $^{181}\text{Ta}^{16}\text{O}^+$, are listed.

Frequency	$^{21}\text{Ne}^+$	$^{85}\text{Rb}^+$	$^{133}\text{Cs}^+$	$^{181}\text{Ta}^{16}\text{O}^+$
$\omega_-/2\pi$	1.083 kHz	1.083 kHz	1.083 kHz	1.083 kHz
$\omega_z/2\pi$	107 kHz	54 kHz	43 kHz	35 kHz
$\omega_+/2\pi$	4.337 MHz	1.071 MHz	684 kHz	461 kHz
$\omega_c/2\pi$	4.338 MHz	1.072 MHz	685 kHz	462 kHz

Now, that the situation of the three eigenfrequencies is illustrated, the classical and the quantum mechanical relations, to describe the stored total energy of an ion in a Penning trap, can be introduced.

$$E_{tot} = E_{kin} + E_{pot} = E_+ + E_- + E_z, \quad (2.21)$$

where the energies are corresponding to their eigenmodes, respectively. The particles time-averaged kinetic and potential energies are [Maj06]:

$$\bar{E}_{kin} = \frac{1}{2}m\omega_+^2\rho_+^2 + \frac{1}{2}m\omega_-^2\rho_-^2 + \frac{1}{4}m\omega_z^2z_0^2, \quad (2.22)$$

$$\bar{E}_{pot} = \frac{1}{4}m\omega_z^2(z_0^2 - \rho_+^2 - \rho_-^2). \quad (2.23)$$

For the sake of clarity, the contribution of each eigenmotion is summarized in the following:

$$E_+ = \frac{m}{2}\omega_+^2\rho_+^2 - \frac{m}{4}\omega_z^2\rho_+^2, \quad (2.24)$$

$$E_- = \frac{m}{2}\omega_-^2\rho_-^2 - \frac{m}{4}\omega_z^2\rho_-^2, \quad (2.25)$$

$$E_z = \frac{m}{4}\omega_z^2z_0^2 + \frac{m}{4}\omega_z^2z_0^2. \quad (2.26)$$

Here, $\rho_\pm$ and z_0 determine the amplitudes of the eigenmotions. Each energy is separated into two parts. The first part determines the kinetic energy, whereas the second part the potential energy contribution.

On condition, as it is shown in Eq. (2.26), the energy of the axial motion oscillates between the kinetic and the potential energy. In the case of the reduced cyclotron motion ω_+ , the kinetic energy component outweighs due to the relation $\omega_+ \gg \omega_z$, as can be seen in Eq. (2.24), while the ω_- -motion pinpoints the potential energy of the harmonic system, caused by the fact that $\omega_z \gg \omega_-$, as described in Eq. (2.25). Hence, a bigger reduced cyclotron motion radius ρ_+ enhances the kinetic radial energy of the stored particles, whereas an enhancement of the magnetron radius ρ_-

reduces the magnetron potential energy, stored in the radial plane. By virtue of this effect a dissipative process, like collisions with rest gas, leads to an augmentation of ρ_- and after a certain moment to a loss of the stored particles. Thus, it is obvious that an environment with high-vacuum is necessary in the precision Penning trap. However, the same effects play a major role in the buffer-gas cooling processes in the preparation Penning trap and will be explained in Section 2.2.5. Since in an ideal trap the ion has the eigenfrequencies decoupled, the energy of a particle can also be described as a quantum harmonic oscillator [Kre92]. The motions in the radial-plane describe a quantum two-dimensional isotropic harmonic oscillator, whereas the axial-motion is characterized by an one-dimensional harmonic oscillator. The quantum mechanical energy levels, which increase or decrease the total energy of the system are called *Landau levels*⁷.

$$E_{tot} = \hbar\omega_+ \left(n_{\omega_+} + \frac{1}{2} \right) - \hbar\omega_- \left(n_{\omega_-} + \frac{1}{2} \right) + \hbar\omega_z \left(n_{\omega_z} + \frac{1}{2} \right), \quad (2.27)$$

whereas n_{ω_+} , n_{ω_-} , n_{ω_z} , are the principle quantum numbers. At this juncture, one notices that the magnetron energy has a negative sign as shown in Eq. (2.27), derived by the solution of Eq. (2.15). In the following Fig. 2.7 this effect is visualized in a quantum mechanical picture with discrete values of the three-dimensional harmonic oscillator.

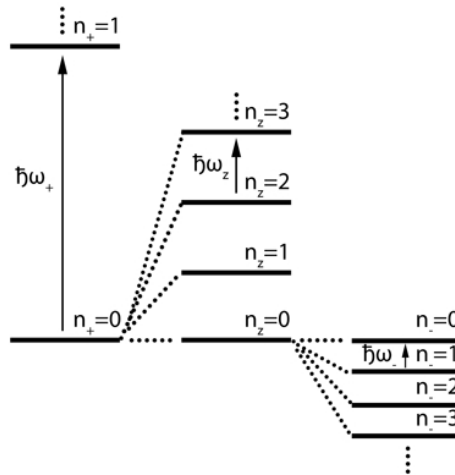


Figure 2.7: The splitting of the Landau levels described by harmonic oscillators for spinless charged particles for an ideal Penning trap. The distance of the three independent harmonic oscillators, the reduced cyclotron ω_+ , the axial ω_z , as well as the magnetron frequency ω_- are not true to scale. The corresponding quantum numbers are represented by n_+ , n_z and n_- , as shown in [Geo09].

⁷Here, considering the nonrelativistic case of a spinless particle, confined in a time-independent electromagnetic field.

Real Penning traps

Penning traps are not ideal as discussed in the previous Section 2.2.3. Reasons for this include the finite size, the opening and imperfection of the trap's electrodes and the inhomogeneity of the magnetic field. Some of these imperfections can evolve with time, due to pressure and temperature changes. These real conditions affect the electric and magnetic field and with this the measured ion eigenfrequencies can become shifted.

• Electric Field Imperfections

The effect of the electric field imperfection is due to the facts, obvious from Fig. 2.6, e.g. that the ring of the trap is segmented to ensure driving the ion's motion through variable external rf-fields, as well as the end caps exhibit apertures for the injection and ejection of the charged particles. These points lead to a deviation from the pure quadrupole field distribution, as previously discussed in case of the ideal Penning trap. To reduce this aperture effects, correction electrodes are added to the trap. These external corrections reduce higher-order potential distortions. The possibility to carry out these adjustments very precisely resulted in a choice of the cylindrical design of modern Penning traps [Muk08; Ket14], giving in addition the constructive freedom at electrode manufacturing. The trap potential can be expressed, with its coefficients C_k under consideration of the Legendre polynomials $P_k(\cos \theta)$ to:

$$\Phi(r, z) = \frac{U_{trap}}{2} \sum_{k=1}^{\infty} C_k \left(\frac{\rho}{d} \right)^k \cdot P_k(\cos \theta), \quad (2.28)$$

written up to the sixth-order derivative as:

$$\begin{aligned} \Phi(r, z) = \frac{U_{trap}}{2} & \left[\frac{C_2}{d^2} \left(z^2 - \frac{r^2}{2} \right) + \frac{C_4}{d^4} \left(z^4 - 3z^2 r^2 + \frac{3}{8} r^4 \right) \right. \\ & \left. + \frac{C_6}{d^6} \left(z^6 - \frac{15}{2} z^4 r^2 + \frac{45}{8} z^2 r^4 - \frac{5}{16} r^6 \right) + \dots \right]. \end{aligned} \quad (2.29)$$

The odd terms can be neglected, as a result of the mirror symmetry in z -direction. In an ideal trap $C_2 = 1$, whereas all other terms are $C_k = 0$, for $k > 2$, if the ions are stored in the homogeneous ideal trap region, $\rho \ll d$. All additional higher-order terms contribute to a frequency shift of the measured cyclotron frequency [Gab09]. The potential terms C_4 and C_6 are considered as perturbations of the quadrupole field and can be written as an influence on the ions' sum frequency derived from the first-order perturbation theory

[Maj06; Ket14] to:

$$\Delta\omega_c^{el} = \frac{3}{4} \frac{\omega_z^2}{\omega_+ - \omega_-} \left[\frac{C_4}{d^2} (\rho_-^2 - \rho_+^2) + \frac{5}{2} \frac{C_6}{d^4} [3z_0^2 (\rho_-^2 - \rho_+^2) + (\rho_+^4 - \rho_-^4)] + \dots \right], \quad (2.30)$$

for the fourth-order and the sixth-order, i.e. counting for the octupole C_4 and dodecapole C_6 contributions. Here, ρ_- is the radius of the magnetron motion, where ρ_+ is the radius of the reduced cyclotron motion. That the frequency shift $\Delta\omega_c^{el}$ is in the first order mass-independent can be shown with the previously introduced influences of the cyclotron frequency in Eq. (2.30) by extending the first coefficient with the magnetron frequency ω_- using Eq. (2.16a):

$$\frac{\omega_z^2}{\omega_+ - \omega_-} = \frac{2\omega_-}{1 - \omega_-/\omega_+} \approx 2\omega_- \approx \frac{U_{trap}}{2d^2 B^2}. \quad (2.31)$$

The E-field perturbations can be minimized by use of correction electrodes, which will be shown in Section 4.4.3. Another not to be underestimated effect is the relative deviation of the electrostatic field with respect to the magnetic field axis, if both are not completely perpendicular to each other. This results in a shift of the sum frequency ω_c about the factor calculated by [Gab09]. The systematic frequency shift can be shown to be mass-independent, with respect to small tilt angles, $\Theta \ll 1$ and small ellipticity $\epsilon \ll 1$ to:

$$\Delta\omega_c^{tilt} \approx \omega_- \left(\frac{9}{4} \sin^2 \Theta - \frac{1}{2} \epsilon^2 \right). \quad (2.32)$$

The angle Θ at ISOLTRAP is minimized as low as $\Theta = 0.08^\circ$. The results and the method of alignment of ISOLTRAP's vertical beam line by means of a collimated electron gun positioned at the center of the magnetic field, was accomplished in 2013 and is well documented in [Ata16].

• Magnetic Field Imperfections

Due to the fact that the mass is determined by the magnetic field among other things, imperfections and a lack of stability lead to incorrect mass measurements. Nowadays, a typical homogeneity in the order of $\Delta B/B < 10^{-7}/\text{cm}^3$ can be achieved. The temporal stability of the magnetic field at ISOLTRAP is $\delta B/\delta t \cdot 1/B \sim 6.4 \cdot 10^{-10}/h$ [Ata16], competing with the currently most stable magnet in the world, which is in the order of $\delta B/\delta t \cdot 1/B < 17 \cdot 10^{-12}/h$ [Dyc99]. The following contributions, affecting the stability and homogeneity of the magnetic field the most: First of all the finite size of the coils of the magnet, limiting the volume of stability. In addition, the quality of the used materials in the trap or the surroundings in vicinity of the magnet is of importance. Materials with magnetic susceptible impurities distort the magnetic

field⁸. In order counteract this, the electrodes and the holder of the trap are made of oxygen-free, high-conductivity copper and glass ceramics. To ensure the homogeneous magnetic field, the magnet is equipped with a series of shim coils which generate a secondary magnetic correction field by setting the correct value of an electrical current. The systematic frequency shifts that can arise from the magnetic field are well described in [BG86]. The lowest order correction, which affects the cyclotron frequency, can be derived from the magnetic hexapole component of the magnetic field to:

$$B = B_0 + B_2 \left(z^2 - \frac{r^2}{2} \right), \quad (2.33)$$

Here, B_0 represents the magnetic field strength at the center of the Penning trap, where the relative strength of the hexapole component is given by B_2 . The correction for the cyclotron frequency is then determined by:

$$\Delta\omega_c^{mag} \approx B_2\omega_c \left(z^2 - \frac{r^2}{2} \right). \quad (2.34)$$

Further contributions are temperature and pressure fluctuations, which influence the temperature-dependent component impurities of the materials. These effects are discussed in Section 4.4.

For the correction procedure and the results of the electric and magnetic field optimization the Penning-trap mass spectrometer is well described in [Muk08; Bec09].

- **Ion-Ion interaction**

Space-charge effects occur in presence of more than one ion in the trap. These effects lead to shifts of the eigenfrequencies, as a result of the additional Coulomb field, which superimposes the harmonic-trapping net-potential. The extensive studies on these effects with Penning traps are summarized in [Bol94; Por15]. In relation to the eigenfrequencies of a single ion, ions of the same mass-to-charge ratio m/q have a factor of 7.5 smaller deviation of the eigenfrequencies, compared to ions with different mass-to-charge values [WA07]. The cyclotron frequencies are shifted to lower values, resulting in a heavier apparent mass the higher the mass difference between the two ion species in the trap is, while the magnetron frequency value increases. The shift of the eigenfrequencies is proportionally affected by the amount of ions in the trap.

- **Mirror charges**

Another frequency shift effect can be caused by mirror charges, which are induced by the charge of the ion into the electrodes, leading to a perturbation

⁸Leading to the so-called flux creep effect [And62; AK64].

of the harmonicity of the trap potential. The electric field of the resulting image charge is approximately:

$$\mathbf{E}_{mir} = \frac{1}{4\pi\epsilon_{per}} \frac{qr_0}{(r_0^2 - r^2)^2} \mathbf{r}. \quad (2.35)$$

Where ϵ_{per} is the absolute permittivity. This additional electric field causes a frequency shift of the axial eigenfrequency by:

$$\Delta\omega_z^{el} = -\frac{1}{4\pi\epsilon_{per}} \frac{q^2}{2mr_0^3\omega_z}. \quad (2.36)$$

For a detailed review, see [Por01].

2.2.4 Penning traps working principle

In the following, various ion excitation schemes will be presented. These schemes can be used to increase either the amplitude of a certain radial motion, or convert from one motion into another by coupling two eigenmodes, as in the case of the coupled harmonic oscillator. In addition, the order in which each radio-frequency (rf) scheme will be applied to the trap electrodes is shown.

Dipole Excitation

Applying a dipole excitation on the ring segments of the Penning trap, manipulates just one specific eigenmotion at a time. This dipole excitation transfers energy from the frequency-generator into the system. Such an excitation in case of the magnetron frequency $\omega_{rf} = \omega_-$, which is in first-order mass independent, as shown in Eq. (2.20), will affect ions of all species. To manipulate only the radius of a certain specific mass, a dipole excitation with $\omega_{rf} = \omega_+$, or $\omega_{rf} = \omega_z$ as an axial dipole frequency excitation applied to the end caps, has to be used. The phase difference for dipole excitation between two opposite electrodes must be 180° , as shown in Fig. 2.8 a).

The generated $\mathbf{E}$ -fields behave as follows with a dipole excitation at $\omega_{rf} = \omega_\pm$, for the radial eigenmodes:

$$\mathbf{E}_{dip} = \frac{U_{dip}}{d} \cdot \cos(\omega_{rf}t - \Phi_{rf}) \cdot (d_x \cdot \hat{\mathbf{e}}_x + d_y \cdot \hat{\mathbf{e}}_y), \quad (2.37)$$

where U_{dip} indicates the amplitude of the radio-frequency voltage and d , as previously mentioned in Eq. (2.8), describes the characteristic trap dimension.

While the $\mathbf{E}$ -field for the axial eigenmotion by use of $\omega_{rf} = \omega_z$ results to:

$$\mathbf{E}_z = \frac{U_{dip}}{d} \cdot \cos(\omega_{rf}t - \Phi_{rf}) \cdot z \cdot \hat{\mathbf{e}}_z. \quad (2.38)$$

The dipolar excitation, either at the reduced cyclotron frequency ω_+ or the magnetron frequency ω_- , can be used to change the motion's radius. The solution of the

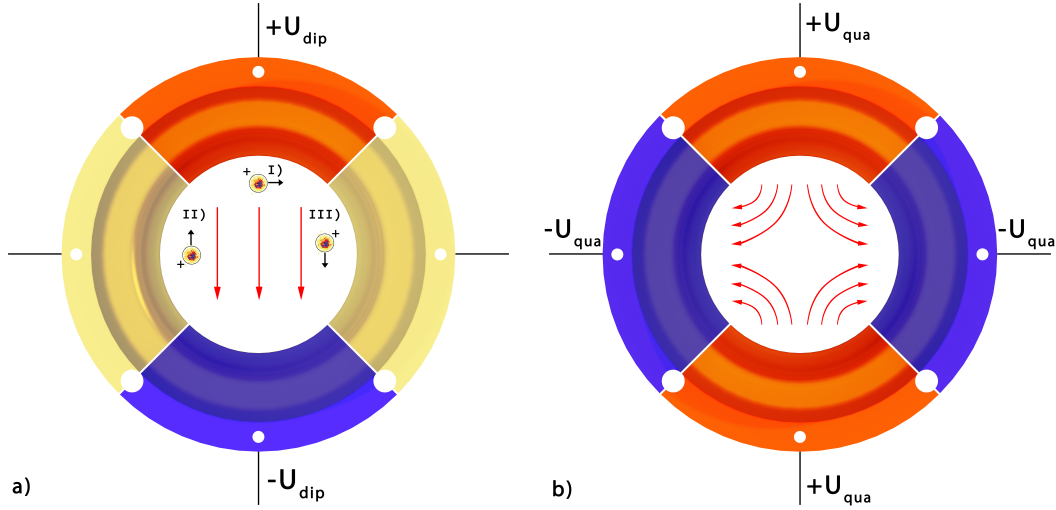


Figure 2.8: The ring electrodes are shown from a top view into the trap. The left hand side a) shows the electrodes with the applied radio-frequency pulse for a dipolar excitation. Depending on the ions' motion phase, it is possible to enhance the energy of the system with $\Delta\Phi = 0$ ion position I) or respectively, with $\Delta\Phi = \pi/2$ at position II). By applying a dipole pulse with a shift of $\Delta\Phi = 3\pi/2$, as shown with ion position III), the energy in the system can be even reduced under certain circumstances. On the contrary side, b) represents the electrode configuration for the quadrupole excitation scheme.

ion motion can be obtained, as described in [Bla03; Kre08], by use of the Laplace transformation and the absolute value of the their velocity vector:

$$\lim_{\Delta\omega \rightarrow 0} r_{\pm}(t_{dip}) = \sqrt{r_{\pm}^2(0) + \left(\frac{qU_{dip}}{md} \frac{t_{dip}}{2(\omega_+ - \omega_-)} \right)^2} \mp \frac{r_{\pm}(0)qU_{dip} \sin(\Delta\Phi)}{md(\omega_+ - \omega_-)^2} t_{dip}, \quad (2.39)$$

here, $\Delta\omega = \omega_{rf} - \omega_{\pm}$ and $\Delta\Phi_{\pm} = \Phi_{rf} - \Phi_{\pm}$. With the motion's behavior the phase dependence between the excitation phase and the localization of the ions is of importance and will influence the radius differently, as described in the following three scenarios for the magnetron motion, corresponding to the positions in Fig. 2.8 a) I) - III):

- $\Delta\Phi_- = 0$: A slowly grow of the magnetron radius ρ_- at the beginning can be obtained before a linearly increasing trend is pursued.
- $\Delta\Phi_- = \pi/2$: In this case the dipolar excitation radio-frequency is anti-parallel to the ion's motion. The magnetron radius ρ_- is increased right from the beginning by the excitation and follows a linear trend.
- $\Delta\Phi_- = 3\pi/2$: This scenario describes parallel orientation of the ion's motion and the dipolar excitation radio-frequency field. In resonance with the magnetron motion $\omega_{rf} = \omega_-$, the magnetron radius ρ_- decreases at the beginning linearly with time until a certain moment, where it increases again linearly. This procedure can be used to reduce an initial magnetron motion.

The aforementioned phase dependence is illustrated in the following Fig. 2.9.

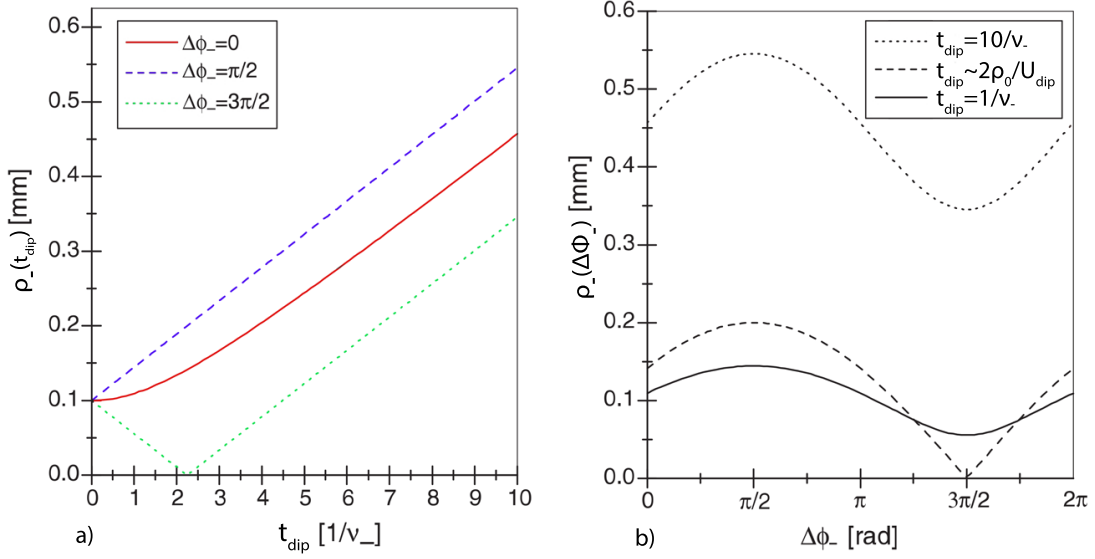


Figure 2.9: Figure a) shows the phase dependence of the dipolar radio frequency field on the magnetron ion motion as a function of the duration time t_{dip} of the excitation pulse. The three different scenarios correspond to the differential phase $\Delta\Phi_-$ between the rf-pulse and the initial magnetron phase. The reduction of the magnetron radius as in the case of $\Delta\Phi_- = 3\pi/2$ (dotted line) is favored to reduce the initial magnetron radius ρ_0 . In b) the magnetron radius ρ_- is plotted as a function of the differential phase between the rf excitation and the initial magnetron phase. Three different dipole excitation-duration times t_{dip} are shown, as mentioned in [Bla03; Kre08].

The sine-like structure in Fig. 2.9 b), results from the sine-term in Eq. (2.39).

Quadrupole Excitation

To convert one ion motion into another, a quadrupole excitation has to be used to allow the coupling between two eigenmodes. For the quadrupole excitation, four ring electrodes at ISOLTRAP are supplied by a rf signal, as presented in Fig. 2.8 b), which generates the following E-field:

$$\mathbf{E}_x = -\frac{U_{qua}}{2d^2} \cdot \cos(\omega_{rf}t - \Phi_{rf}) \cdot y \cdot \hat{\mathbf{e}}_x, \quad (2.40)$$

$$\mathbf{E}_y = \frac{U_{qua}}{2d^2} \cdot \cos(\omega_{rf}t - \Phi_{rf}) \cdot x \cdot \hat{\mathbf{e}}_y. \quad (2.41)$$

Here U_{qua} is the amplitude of the rf excitation applied in a 90° phase shift to the four-fold-segmented ring electrodes.

Solving the equation of the periodical quadrupole excitation of the ion's motion in

a Penning trap, as described in [Kön95], leads to:

$$r_{\pm}(t) = \left(r_{\pm}(0) \cos(\omega_R t) \mp \frac{1}{2} \frac{r_{\pm}(0) i(\omega_{rf} - \omega_c) + r_{\mp}(0) k_{rf}^{\pm}}{\omega_R} \cdot \sin(\omega_R t) \right) e^{i \frac{1}{2} (\omega_{rf} - \omega_c) t}, \quad (2.42)$$

where $\omega_R = \frac{1}{2} \sqrt{(\omega_c - \omega_{rf})^2 + k_{rf}^2}$ and $k_{rf}^{\pm} = k_{rf} e^{\pm i \Delta \Phi}$, $k_{rf} = \frac{q}{m} \frac{U_{qua}}{2d^2}$.

In the following the step of the conversion from the energy perspective will be clarified. The stored radial kinetic energy E_{rad} for a quadrupole excitation can be written as:

$$E_{rad} \propto \omega_+^2 r_+^2(t) + \omega_-^2 r_-^2(t) \approx \omega_+^2 r_+^2(t), \quad (2.43)$$

where the increase during the conversion can be written in the form of the normalized energy gain to [Muk08]:

$$E_{rad} = \frac{4g^2}{\omega_R^2} \cdot \sin^2 \frac{\omega_R^2 T_{conv}}{2}. \quad (2.44)$$

The resonant frequency, which describes the beating between the conversions from one motion into another [Bol90], is the so-called Rabi-frequency $\omega_R = \sqrt{(2g)^2 + \delta_{dt}^2}$. The coupling factor g is given by:

$$g = \frac{q}{4m(\nu_+ - \nu_-)} \cdot \frac{U_{qua}}{d^2}. \quad (2.45)$$

Here $\delta_{dt} = 2\pi(\nu_{rf} - \nu_c)$ is the frequency detuning between the excitation frequency from the rf-generator and the resonance frequency ω_c .

One full motion conversion lasts T_{conv} , which pursues physically the period:

$$T_{conv} = \frac{m}{q} \cdot \frac{2d^2}{U_{qua}} (\omega_+ - \omega_-) \pi = \frac{B}{\omega_c} \cdot \frac{2d^2}{U_{qua}} (\omega_+ - \omega_-) \pi. \quad (2.46)$$

An over-conversion exceeding the time period T_{conv} would convert the cyclotron motion back into magnetron motion, whereby the gained excess of energy is drained back into the frequency generator. A similar behavior as in a LC-circuit.

The method of the interconversion between the motions is used for different needs. For example the buffer-gas cooling technique (Section 2.2.5) or different types of detection techniques, e.g., determination of the ion time of flight, as shown in Section 2.2.6 or the novel phase-imaging technique, explained in Chapter 4.

2.2.5 Preparation Penning trap

After the bunched ion beam is transferred from the MR-ToF MS, as described in Section 2.2.2, the ion reaches the second pulsed decelerating cavity. This second

decelerating cavity, located in Fig. 2.3 between No. 5-6, reduces the kinetic energy of the ion bunch to a value of $E_{kin} \leq 100$ eV before injecting the ions into the preparation Penning trap (also called lower trap or LT). The preparation Penning trap is an important prior stage for high-precision mass measurements. To prepare the ions at ISOLTRAP, the mass selective buffer-gas cooling technique is used [Sav91], which was already introduced briefly in Eq. (2.1). The preferred buffer-gas that enables the cooling process at ISOLTRAP is usually Helium (He). Its high ionization potential prevents the ion to be cooled from passing through a charge exchange during the cooling process, thus preventing charge neutralization and loss of the ion. The viscous drag approach described previously in Eq. (2.2) can be used to describe the evolution of the magnetron and cyclotron motion with a dissipative parameter by:

$$\rho(t) = U_\rho \cdot e^{-\gamma_\pm t}. \quad (2.47)$$

Where U_ρ is the initial amplitude of the corresponding motion and $\gamma_\pm$ represents the damping constant for the radial motions described as:

$$\gamma_\pm = \pm \delta \frac{\omega_\pm}{\omega_+ - \omega_-}. \quad (2.48)$$

In the RFQ Section 2.2.1, it was mentioned that the charged particle is trapped in a 3D pseudo-potential field, where collision and dipole interaction with the He buffer-gas simultaneously reduces the energy in all motions and as a result by the demand of smaller phase space volume, the ion gets centered in a potential well. However, a trapped charged particle in an electrostatic magnetic field behaves differently. On the one hand, the manipulation of the three decoupled ion motions becomes possible, on the other hand, each motion is connected to different potentials, which explains why the buffer gas cooling in a Penning trap has to take place differently than in a Paul trap, such as the RFQ. As the three eigenmotions couple differently to the axial quadrupole electrostatic trapping potential and the axial static magnetic field, the following picture Fig. 2.10 describes intuitively the motions and their behavior.

At the beginning of this section, it was shown in Eq. (2.13) that the axial motion does not feel the azimuthal quadrupole field in an ideal Penning trap. The effect which occurs if the axial motion interferes with a buffer-gas that centers the ion, while it loses energy, can be visualized with the energetic behavior in Fig. 2.10 a). The radial motions were introduced in Eq. (2.15) and are splitted into two components, the slow magnetron motion and the fast cyclotron motion. The cooling ability of the latter motion with buffer-gas interaction can be seen in Fig. 2.10 b). Therefore, the motion is favored in the preparation trap for cooling purposes. The reduced cyclotron motion, as well as the axial motion are accordingly called stable motions. Whereas the magnetron motion behaves differently due to the electrical repulsion field. This field drives the $\mathbf{E}_{rad} \times \mathbf{B}$ drift, which reduces the potential energy by enhancing the magnetron radius, with every dissipative process. This effect results into the potential hill, illustrated in Fig. 2.10 c). By the fact that the magnetron motion is slow compared to all other motions and that the velocity vector

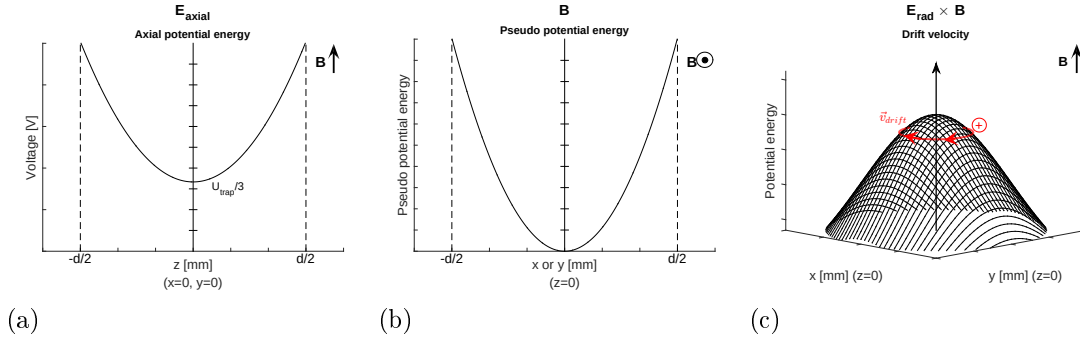


Figure 2.10: The three decoupled eigenmotions of a charged trapped particle in an axial electrostatic quadrupole trapping potential, superimposed by an axial static magnetic potential. a) Describes the axial motion with its frequency ν_z , as a one-dimensional harmonic oscillator with its minimum at $z = 0$. b) Shows the resulting pseudo potential $\mathbf{B}_{axial}$ of a cyclotron motion, with its minimum at the center of the radial plane, which indicates the cooling capability. c) Illustrates the magnetron motion, whereas the center of the gyrating charged particle (i.e. the guiding center) experiences the $\mathbf{E}_{rad} \times \mathbf{B}$ drift, with its velocity vector $\mathbf{v}_{drift}$, representing the metastable trajectories on the potential energy slope. If a dissipative process occurs the magnetron radius of an ion gets increased, while the potential energy stored in the ion gets reduced.

$\mathbf{v} = c \cdot \mathbf{E} \times \mathbf{B} / B^2$ moves perpendicular to the electric and magnetic field $\mathbf{E} \perp \mathbf{B}$, the motion moves almost unimpeded in a real Penning trap. These conditions acting as a velocity filter. Therefore, with almost years of half-life⁹, the magnetron motion is defined as a metastable motion [BG86]. It also shows that in a precision Penning trap, the better the vacuum, the better the control of the magnetron motion. For further details see Section 2.2.6.

The previous picture in Fig. 2.10 b) and c) can be explained with the two stability conditions in the Penning trap for the magnetron motion and the cyclotron motions. The first stability criteria refers to the magnetron motion, by raising the question at which velocity the electrostatic force $\mathbf{F}_{el}$, pointing outward directed with respect to the Penning trap center, is equal to the inward directed Lorentz force $\mathbf{F}_{lor}$. Both forces were introduced in Eq. (2.5). The following condition fulfills the required equilibrium:

$$\begin{aligned} \mathbf{F}_{lor} &= \mathbf{F}_{el}, \\ q \cdot \mathbf{E} &= q \cdot \mathbf{v} \times \mathbf{B}. \end{aligned} \quad (2.49)$$

The second stability condition refers to the cyclotron motion. Here the question arises when the difference between the Lorentz force and the electrostatic force,

⁹By use of an ultra-high vacuum.

which determines the centrifugal force $\mathbf{F}_{cp}$, is the prerequisite for a circular motion:

$$\begin{aligned} \mathbf{F}_{lor} - \mathbf{F}_{el} &= \mathbf{F}_{cp}, \\ q \cdot \mathbf{v} \times \mathbf{B} - q \cdot \mathbf{E} &= \frac{m\mathbf{v}^2}{\mathbf{r}}. \end{aligned} \quad (2.50)$$

If a dissipative interaction takes place, the velocity vector on the right hand side of Eq. (2.50) is faster reduced, due the quadratic term, than the velocity vector of the Lorentz force on the left hand side. This describes that the increase of the magnetron radius is slower, than the decrease of the cyclotron-motion radius. This behavior favors the conversion into the reduced cyclotron motion to reduce the magnetron radius of a certain ion species by buffer-gas cooling in the preparation Penning trap.

Now, as the interaction with the trapping potentials and the damping processes is clarified, a more detailed view of the ISOLTRAP trapping scheme is possible. Note that without a driving rf-field (azimuthal) a trapped, charged particle, surrounded by buffer gas, loses its axial and cyclotron motion, whereas the magnetron radius increases. Thus, the ion would be lost after a certain moment when it hits the ring electrodes. An example of this procedure is shown in Fig. 2.11 a). To compensate for this behavior, a driving quadrupole rf-field is required that controls the sideband conversion of the magnetron motion into the reduced cyclotron motion. This process pumps the kinetic energy of the magnetron motion into the cyclotron motion, as described in Eq. (2.42). This leads to a higher reduced cyclotron velocity $\mathbf{v}_+$ which increases its radius ρ_+ . Since a cyclotron motion gets cooled through buffer-gas interaction, unlike the magnetron motion, the ions lose their potential and kinetic energy, which leads to a centering of the ions. The correct damping regime at ISOLTRAP can be achieved by scanning the conversion and trapping times at a fixed buffer-gas pressure. The resulting ion trajectories are shown in Fig. 2.11 b).

This procedure can be used as a mass-selective buffer-gas filter. Prior the MR-ToF MS, as an isobaric purifier at ISOLTRAP, the LT was the state-of-the-art tool for that purpose.

The ejection of the ion bunch from the MR-ToF MS initiates the preparation Penning trap cycle. After the ions are trapped, their initial axial motion is too large. Thus, the motion has to be cooled by buffer-gas interaction before any other radio-frequency is applied to the ring electrodes in the preparation Penning trap. This axial cooling is a waiting time of ~ 50 -250 ms in which the ion bunch is held in the buffer-gas, to reduce its axial motion. A short and broad quadrupole rf ω_{rf} excitation is sent to the ring electrodes to reduce possible initial magnetron motions by converting them into the reduced cyclotron motion. After all motions are pre-cooled, the initial starting conditions are set to apply a dipole excitation on the magnetron frequency $\omega_{rf} = \omega_-$. Now all ions, including potential contaminant masses, circle around the center of the trap on the same magnetron radius. In a subsequent step, a quadrupole rf at the true cyclotron frequency $\omega_{rf} = \omega_c$ is applied. The selective

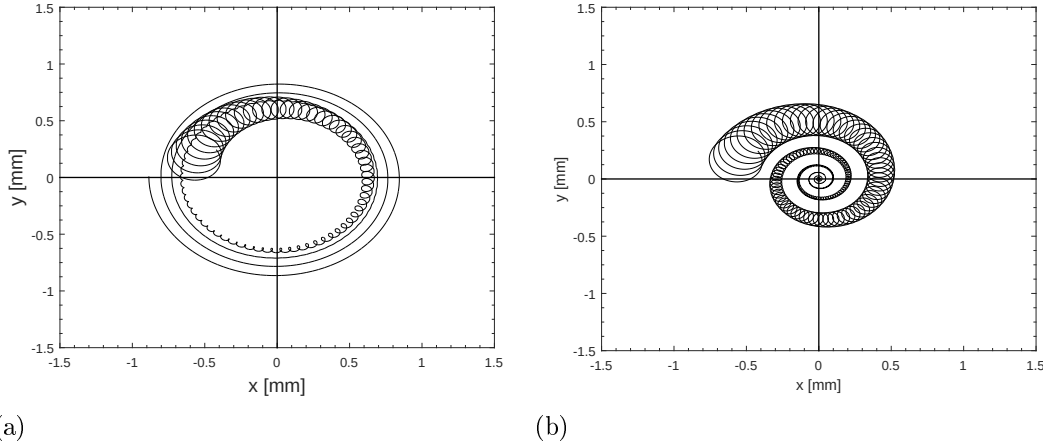


Figure 2.11: The theoretical trajectory of $^{133}\text{Cs}^+$ in the preparation Penning trap. On the left side a), the damping effect without a quadrupole excitation is shown. Here, the cyclotron motion is cooled and the magnetron motion radius gets magnified, an effect that is not desired. b) Illustrates the mass-selective centering with the right cooling time, as well as the correct quadrupole excitation time. These criteria are fulfilled for the so-called critical damping regime. The buffer-gas damping reduces the cyclotron motion, respectively, the magnetron motion in form of sideband cooling, simultaneously. To visualize both effects, the frequency of the motion and the damping parameter are enhanced and are not to scale.

buffer-gas cooling acts and only the species of interest gets centered, while possible contaminants are still orbiting on the magnetron radius. Subsequently, the cooled and centered ion bunch, will be injected into the precision Penning trap. The remaining non-centered possible contaminants aren't able to pass the ejection aperture of the trap of 1.5 mm.

2.2.6 Precision Penning trap

After the ions are ejected from the preparation Penning trap, they are injected into the $B = 5.9$ T precision Penning trap. The axial energy, which is already reduced in the preparation Penning trap, can be controlled with the end cap capture pulse of the precision Penning trap. The time of flight between both traps is mass dependent, therefore, the capture pulse of the precision Penning trap has to be scanned, which will be shown in Section 4.4.1. For this procedure the ring electrodes of both traps have to be at the same potential to minimize the initial axial energy. After the right injection into the trap center, the magnetron dipole excitation is applied to induce energy into the magnetron motion, as described in Section 2.2.4. The magnetron radius at ISOLTRAP has usually the value of $\rho_- = 0.6$ mm. This radially stored energy is converted from the magnetron motion into the pure cyclotron motion with a quadrupole excitation on the right free cyclotron frequency $\omega_{rf} = \omega_c$, as mentioned in Eq. (2.42). The precision Penning trap has an ultra high-vacuum with at least $p < 10^{-8}$ mbar [Bol90], therefore, the collisions with buffer gas are reduced to a

minimum [Geo11]. The behavior without a damping factor is presented in Fig. 2.12.

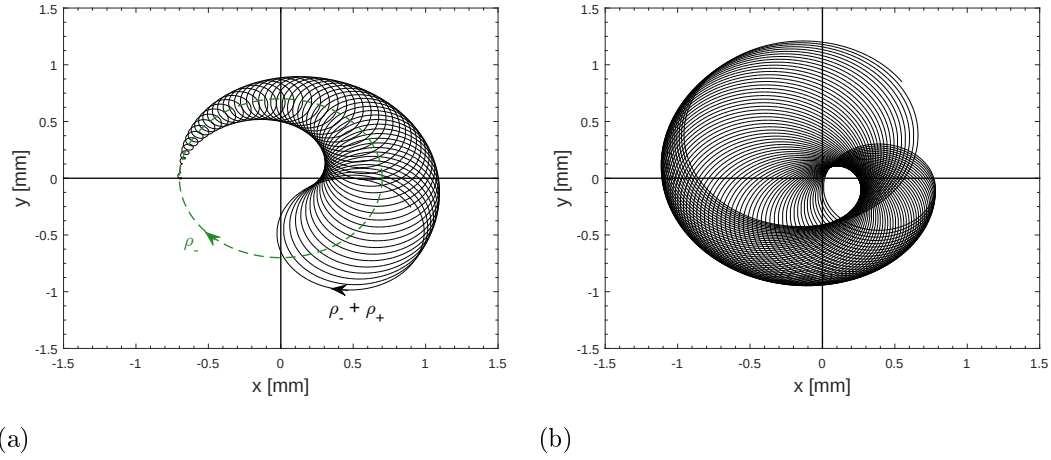


Figure 2.12: Presented are the undamped radial motions for $^{133}\text{Cs}^+$. On the left hand side a) the first part of the conversion with the applied quadrupole radio-frequency $\omega_c = \omega_+ + \omega_-$ is shown. The initial pure magnetron radius ρ_- and corresponding magnetron motion are highlighted in green, while the black marked trajectory shows the cyclotron motion with an increasing amplitude, i.e. increasing ρ_+ . During the conversion, the motion is a superposition of the magnetron and the cyclotron motion ρ_+ and ρ_- . On the right hand side b) an almost fully converted picture of the pure cyclotron motion is visualized; almost none of the initial magnetron motion remains. To visualize the motions, the frequency in both figures was enhanced and is not to scale.

After the full conversion period T_{conv} introduced in Eq. (2.46), the ions orbit still on the same radius as in the case of the magnetron motion on ρ_- , however, on the now converted pure cyclotron motion, with an energy gain of E_{rad} as described in Eq. (2.44). In the last step the ion gets ejected and detected with a destructive detection technique, which will be described in the following.

2.2.7 Various ion detection techniques

Several ways to detect the ion's different motions have been developed. The two most common, as well as a novel technique at ISOLTRAP, as part of this dissertation, are mentioned in the following.

Fourier-Transform Ion-Cyclotron-Resonance Detection Technique

The Fourier-Transform Ion-Cyclotron-Resonance technique (FT-ICR) [MHJ98] traces the ions' motion, while inducing a mirror charge on the electrodes when passing close to them. The signal is amplified differentially and the resumption of the induced mirror charge returns the fingerprint of the frequency of the ions and thus of the mass value. This technique is non-destructive because the ion can be

stored and used over a long period of time. FT-ICR is here just mentioned for the sake of completeness and is not part of ISOLTRAP's detection techniques, due to its requirement of a cryogenic cooled trap and electronics, to reduce thermal noise [Red13; RSB13].

Time-of-Flight Ion-Cyclotron-Resonance Detection Technique

The second method has an over 30 years long history at ISOLTRAP and is the so called Time-of-Flight Ion-Cyclotron-Resonance technique (ToF-ICR) [Grä80]. It is a destructive technique, due to the loss of the ions after each measurement on a micro-channel plate detector (MCP) [Wiz79]. The technique uses the principle that the ion interacts with the magnetic field gradient $\frac{\partial \mathbf{B}}{\partial z}$ via its magnetic moment μ :

$$\mathbf{F} = -\boldsymbol{\mu}(\nabla \cdot \mathbf{B}) = -\frac{E_{rad}}{B} \frac{\partial B}{\partial z} \mathbf{e}_z, \quad (2.51)$$

with:

$$\boldsymbol{\mu} = -\frac{q}{2} (r_+^2 \omega_+ + r_-^2 \omega_-) \mathbf{e}_z. \quad (2.52)$$

The ion gets accelerated due to the gradient force, which is proportional to their orbital magnetic moment and thus their initial radial energy E_{rad} . As already mentioned in Section 2.2.4, all ions are prepared on a certain magnetron radius ρ_- , followed by a conversion to the reduced cyclotron motion by a quadrupole excitation, which leads to an increase of the radial energy in case of resonance. This up-take of energy leads to a stronger coupling of the magnetic moment to the magnetic field lines. This effect ensures the difference in flight times, resulting in the ToF-distribution shown in Fig. 2.13. The corresponding time T_{ToF} that the ions need between the ejection pulse (start trigger) and the MCP detector (stop trigger) determines the time-of-flight of each measurement cycle. The time is given by:

$$T_{ToF}(E_{rad}) = \int_0^{z_{det}} \sqrt{\frac{m}{2(E_0 - qU(z) - |\boldsymbol{\mu}(E_{rad})\mathbf{B}(z)|)}} dz, \quad (2.53)$$

where the integral limits are the center of the trap as a starting point $z = 0$ and the detector position $z = z_{det}$ as the upper limit. E_0 defines the total initial energy of the ion, $U(z)$ and $\mathbf{B}(z)$ are the electrical-potential and magnetic-field distribution, respectively, along the ion's path between the trap center and the MCP detector. Based on the relation Eq. (2.52) it becomes apparent that the coupling to the magnetic-field lines is stronger, the closer the excitation frequency of the ion is to the resonance frequency. It is the correct free cyclotron frequency ω_c , where the conversion is complete, meaning that in Eq. (2.52) ρ_+ has the maximum possible value for the same initial ρ_- , as described previously in Section 2.2.4. The procedure to obtain the typical resonant case, a scan of the frequency ω_{rf} over a broader range is necessary. The free cyclotron frequency ω_c can then be determined from the

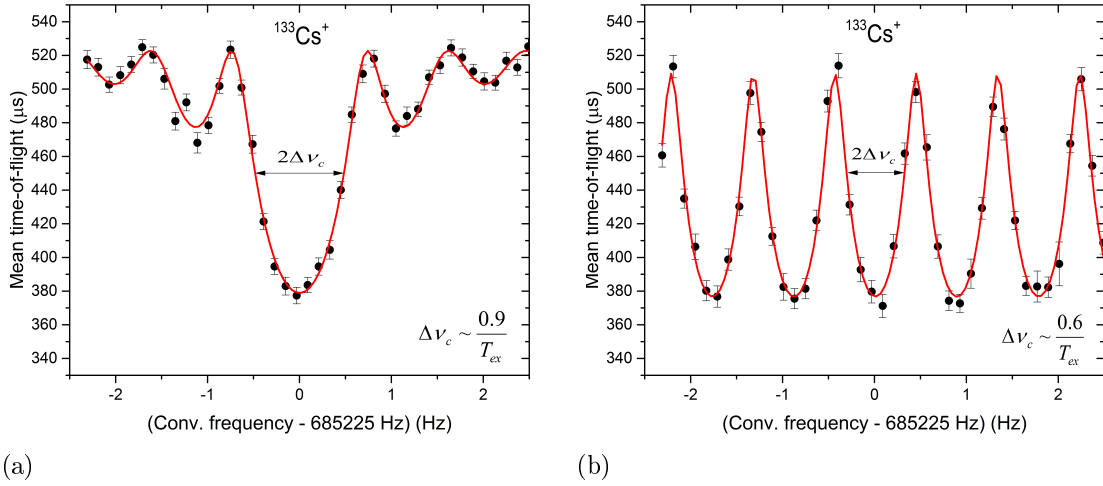


Figure 2.13: a) presents the typical ToF-ICR technique for $^{133}\text{Cs}^+$ with an excitation time of $T_{ex} = 1200$ ms. The ToF of an ion is shorter, the stronger the coupling of its magnetic moment to the magnetic field is. This condition is fulfilled with the resonant quadrupole frequency at $\omega_{rf} = \omega_c$ and the correct conversion time, respectively excitation time $T_{conv} = T_{ex}$ for the given amplitude. In b) the typical Ramsey scheme was applied, where the center fringe represents the cyclotron frequency. The scheme differs from ToF-ICR by the excitation procedure. In ToF-ICR a continuous quadrupole excitation of $T_{ex} = 1200$ ms is applied, whereas in the Ramsey scheme two pulses of T_{ex} are separated by an excitation free waiting time, for 1200 ms as following: $T_{ex} - T_{wait} - T_{ex}$. $T_{ex} = 100$ ms and $T_{wait} = 1000$ ms. The achievable precision $\Delta\nu_c$ is determined by the width of the center fringe. In both pictures the black markers illustrate the experimental values with their ToF uncertainties, where the red solid line shows the fit of the theoretical line shape to the data.

characteristic ToF-ICR spectrum, which is represented in Fig. 2.13, as the center fringe.

In case of Fig. 2.13 a) the response of the standard ToF-ICR-resonance or one-pulse scheme, can be described by the $\text{sinc}^2(x)$ -function. It is the Fourier transform of a rectangular pulse in the time-domain [Bol90; Kön95], which also shows local minima (side-bands), symmetrically distributed around the center¹⁰, respectively, the absolute minimum (resonance frequency). The line width of the center fringe $\Delta\nu_c$ is an indicator of the resolving power $\mathfrak{R}_{ToF-ICR}$:

$$\frac{\Delta m}{m} = \frac{2\pi\Delta\nu_c}{2\pi\nu_c} \approx \frac{1}{\mathfrak{R}\sqrt{N_{ion}}} = \frac{m}{qB T_{ex}\sqrt{N_{ion}}}, \quad (2.54)$$

¹⁰Experimental results [Rin07] and simulations [Kre12] have shown, that the symmetric distribution is only given if the quadrupolar excitation phase Φ_{qua} is either averaged over all phases (free run mode of the frequency generator) or with a fixed phase at $\Phi_{qua} = \pm\Phi_{qua}/2$ with respect to the ion's phase. Latter one is harder to accomplish, therefore, at ISOLTRAP the first solution is persecute. It should be mentioned that even if the distribution is not symmetric, the free cyclotron frequency ν_c is nevertheless correct.

where N_{ion} is the amount of recorded ions in the spectrum. The resolving power for ISOLTRAP after many analyzed data sets [Bol01] is estimated to:

$$\mathfrak{R}_{ToF-ICR} \approx 1.25 \cdot \nu_c \cdot T_{ex}. \quad (2.55)$$

The uncertainty, respectively, the resolving power is therefore Fourier limited by the quadrupole excitation-time T_{ex} ¹¹. In case of the one-pulse standard excitation scheme, the full-width-at-half-maximum (FWHM) defined by the excitation time is $\Delta\nu_c \sim 0.8/T_{ex}$ [Bol01], respectively, $\Delta\nu_c \sim 0.6/T_{ex}$ ¹² for the Ramsey resonance technique [Geo07], assigning the better resolving power to the latter. Due to the multiple fringe structure system response of the Ramsey-type scheme, which makes its use ambiguous in some cases, the single-pulse ToF-ICR is preferred to resolve close lying masses, e.g. isomers.

Extending the information of Table 2.1 with the now obtained mass-resolving power $\mathfrak{R}_{ToF-ICR}$, from Eq. (2.55), one obtains an overview in the following Table 2.2:

Table 2.2: Typical mass resolving power $\mathfrak{R}_{ToF-ICR}$, at the precision Penning trap of ISOLTRAP ($B = 5.9$ T) as an example for singly charged ions, $^{21}\text{Ne}^+$, $^{133}\text{Cs}^+$ and the singly charged molecule $^{181}\text{Ta}^{16}\text{O}^+$, with an observation time of $T_{ex} = 1200$ ms are listed.

Ion	ν_c [MHz]	$\Delta\nu_c$ [Hz]	$\mathfrak{R}$ [10^6]	$N_{ion}[\#]$	$\Delta m/m$	Δm [keV]
$^{21}\text{Ne}^+$	4.338	0.66	6.5	600	6.2×10^{-9}	0.13
$^{85}\text{Rb}^+$	1.072	0.66	1.6	600	2.6×10^{-8}	2.2
$^{133}\text{Cs}^+$	0.685	0.66	1.0	1000	3.2×10^{-8}	4.2
$^{181}\text{Ta}^{16}\text{O}^+$	0.462	0.66	0.69	1200	4.1×10^{-8}	8.2

For the sake of completeness it should be mentioned that the initial cyclotron frequency has to be cooled to its minimum in the preparation Penning trap. It is another limiting factor of the resolving power in the PI-ICR and ToF-ICR technique. Simulations have shown [Kre12] that the width of a ToF-ICR fringe is determined through the initial reduced cyclotron motion. The less energy in the motion, the well determined the width of the fringe will be. Additionally, a proper cooling avoids mixing between the initial magnetron motion and the remaining reduced cyclotron motion in PI-ICR. Thus, a better defined spatial-distribution spot is attainable, as shown in Section 4.4.2.

Before moving on to the detailed description of the newly installed Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique (Chapter 4), as part of the thesis, a brief introduction to the principle of mass determination using Penning traps and the evaluation of masses by use of the ToF-ICR technique (Chapter 3) is shown in the following.

¹¹This indicates the uncertainty limit of the ToF-ICR technique for mass measurements of short-lived isotopes, which creates a limit on the maximum resolving power with respect to their half-lives.

¹²Here, T_{ex} for the two-pulse Ramsey scheme is determined by the total time of the two excitation pulses plus the excitation-free time.

Mass determination at ISOLTRAP

The results from mass measurements with Penning traps, independent with which technique the mass was accomplished, are rather given as frequency ratios with respect to a well known reference mass M_{ref} . Thereby, the reference mass should be as close as possible in mass, to prevent mass dependent frequency shifts, due to electric and magnetic field imperfections, respectively, time-dependent changes, as shown in Section 2.2.3. The atomic mass of the nuclide of interest M_{NoI} can be then obtained by:

$$M_{NoI} = \frac{\nu_{c,ref}}{\nu_{c,IOI}} \cdot (M_{ref} - m_e) + m_e, \quad (2.56)$$

where m_e is the electron mass, M_{ref} the reference mass, as well as the frequency ratio $r = (\nu_{c,ref}/\nu_{c,IOI})$ of the reference ion (*ref*) and the ion of interest (*IOI*).

To determine the mass of an *IOI* at ISOLTRAP, three types of Penning trap measurement schemes are available. The standard ToF-ICR one-pulse scheme, the double pulse Ramsey scheme (Section 2.2.7) and the PI-ICR technique (Chapter 4). In order to compare all three techniques with each other in respect to a mass determination, an example for a mass measurement of ^{129}Cs , as the *IOI*, by use of ^{133}Cs as *ref* ion, with an excitation time¹³ of $T_{ex} = 600$ ms is shown in the following Fig. 2.14:

The grey band in Fig. 2.14 represents one standard mass deviation of ^{129}Cs , given by the AME2016¹⁴ [Wan17]. For each data point, a measurement was performed as described above. With exactly the same excitation, respectively, accumulation time, the difference in uncertainty becomes immediately apparent when comparing all three techniques to each other. The figure 2.14 shows that the standard single-pulse ToF-ICR technique is the least precise determination technique among the mentioned. The improvement in precision and gain in resolving power of the new PI-ICR technique with respect to the former two ToF-ICR techniques will be discussed in more detail in Chapter 4.

The five times more precise measured mass of the ^{129}Cs using the PI-ICR technique will be published elsewhere.

Mass uncertainties at ISOLTRAP

In order to be able to claim the accuracy of a measured mass in an experiment, both, the statistical and systematic uncertainties must be known. The overall uncertainty can be attributed to the following factors:

$$\sigma_{tot} = \sqrt{\sigma_{stat}^2 + \sigma_{mass}^2 + \sigma_{res}^2}. \quad (2.57)$$

¹³In case of PI-ICR the excitation free time is called accumulation time.

¹⁴The atomic mass evaluation, which contains least-squares adjustments to all evaluated and accepted experimental masses.

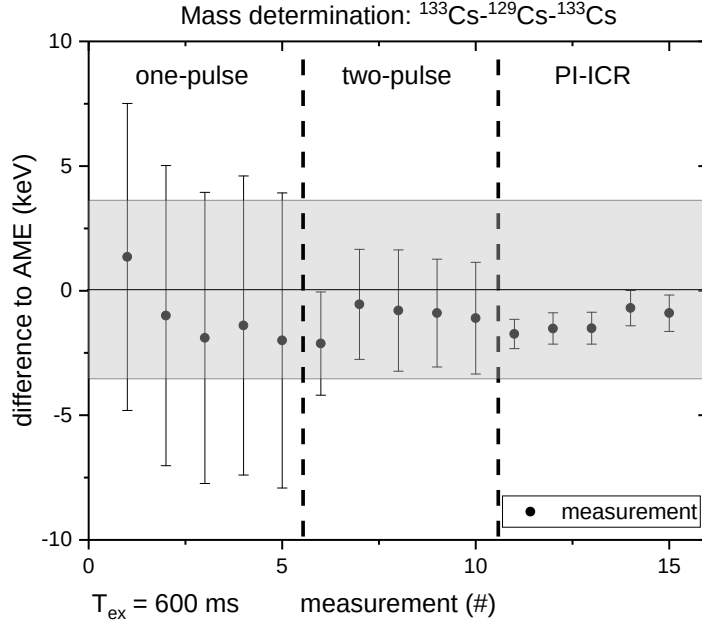


Figure 2.14: Compared are the mass measurements of ^{129}Cs by use of the one-pulse ToF-ICR, the two-pulse Ramsey and the PI-ICR technique with five measurements each. The reference mass used in this measurement was ^{133}Cs . The excitation time, respectively, accumulation time was constant at 600 ms for each technique. The difference in the uncertainty for each technique can be seen. The grey band highlights one standard deviation given by the AME16 [Wan17].

Here, σ_{stat} is the total statistical uncertainty, σ_{mass} the mass-dependent shift and σ_{res} the residual-systematic uncertainties. A more detailed description can be found in [Kel03]. The statistical uncertainties take into account the amount of ions measured and the magnetic field drift that has elapsed over the measurement time of reference ion and ion of interest. Therefore, the statistical uncertainties are based on a compromise between the measurement time and the amount of ions collected.

In comparison, the systematic uncertainties at ISOLTRAP consists of the mass-dependent shift and the residual-systematic uncertainties. In case of the later one, a thoroughly studied mass-independent residual-systematic uncertainty has been determined to 8×10^{-9} , as described in [Kel03]. This value of the residual-systematic uncertainty was used for all mass determinations within this thesis. Due to re-alignment of the whole vertical part of ISOLTRAP in 2013 [Ata16] and the maintenance of the precision Penning trap's magnet, the relative mass-dependent error had to be re-estimated. Prior to the re-alignment, a systematic carbon cluster ion study of different sizes was conducted to determine the mass-dependent shift to $1.6 \times 10^{-10}/u \cdot (m - m_{ref})$. This mass-dependent error occurs due to electric and

magnetic field imperfections in the Penning trap. To date, there has been no carbon cluster study since the re-alignment phase. To be able to determine the mass-dependent shift nevertheless, the mass-dependent uncertainty had to be evaluated by use of the three alkaline ions $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$, which are available from the offline source at ISOLTRAP.

The resulting data is presented in the following Fig. 2.15:

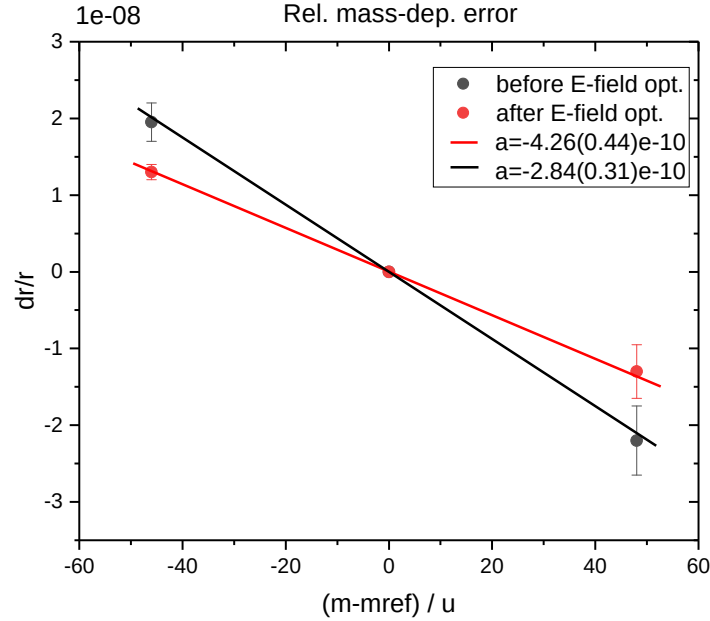


Figure 2.15: Shown are the resulting mass-dependent shifts of ions available at ISOLTRAP for $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$. Two different measurements are presented, one before the electrostatic trapping potential calibration and one after the calibration. The resulting mean average linear trend is used as a mass-dependent shift for all ions to be measured at ISOLTRAP, until a carbon cluster study has been conducted. See text for more info.

For the determination of the mass-dependent shift shown in Fig. 2.15, the ion $^{85}\text{Rb}^+$ has been used as the reference ion, with respect to the mass shifts of $^{39}\text{K}^+$ and $^{133}\text{Cs}^+$, leading to a mass-dependent shift of $3.5 \times 10^{-10} / u \cdot (m - m_{ref})$. In order to obtain a robust value for the mass-dependent shift, the mean value of a non-tuned and a tuned electrostatic field of the precision Penning trap was used for the masses determined within this thesis.

A more detailed description of the masses determined in the context of this work is described in each corresponding chapter.

Chapter 3

Mass measurements with ToF-ICR

In this chapter two measurements will be presented, which were both obtained by use of the standard ToF-ICR technique. The first measurement covers the electron-capture Q_{EC} -value determination for the pair ^{202}Pb - ^{202}Tl Section 3.1. The impact of that pair of nuclides to determine the upper mass limit of the neutrino mass will be discussed. Subsequently, the copper masses $^{75-79}\text{Cu}$, will be presented in Section 3.2, where ^{79}Cu , consisting of $N = 50$ and $Z = 29$, is only one proton above the probable doubly-magic core ^{78}Ni . The influence of the copper masses is emphasized, which explain the configuration of the mass surface only one proton away.

The target material for all two attempted beam times was chosen to be 50 g/cm^2 uranium-carbide, heated to about 2000°C . In case of the production of the neutron deficient ^{202}Pb and ^{202}Tl , the target was directly hit by the 1.4 GeV proton beam. However, to suppress neutron deficient contaminants and enhance the production to the more neutron-rich side, as in the case of the copper $^{75-79}\text{Cu}$ beam time, a tungsten cylinder was used as a neutron converter. In this configuration, the proton beam impinges directly on the neutron converter instead of the target [Kös02]. The subsequent produced secondary neutrons hitting the UC_x target, produce the neutron-rich isotopes by neutron-induced fission [Got14]. Further isotope separation towards the mass spectrometer ISOLTRAP, as well as the cooling and purification processes in the setup, are explained in detail in Chapter 2.

For all two mass measurement series the old detection arrangement behind the Penning trap was used, i.e. before the extraction of the six-way cross, the drift 7 element, as well as the einzel lense, as described in Section 4.3.3. Thus, the thoroughly studied mass-independent error of 8×10^{-9} , as described in [Kel03], was used for all mass determinations within this section. Due to re-alignment of the whole vertical part of ISOLTRAP in 2013, the relative mass-dependent error was re-estimated to be $3.5 \times 10^{-10}/u$, as shown in Section 2.2.7. However, as the carbon cluster study is still pending, the mass-dependent error had to be evaluated by use of the three alkaline ions $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$, which are available from the offline source at ISOLTRAP. Only in the case of the mass ^{202}Pb , which was measured in 2008 at ISOLTRAP [Boe14], the established value of $1.6 \times 10^{-10}/u$ [Kel03]

obtained by carbon cluster measurements was used. As previously described in Section 2.2.3, the magnetic field has an average non-linear drift about factor $\Delta B/t = 6.35 \times 10^{-11} \text{ T/min}$, which is considered in all measurements by their average measurement time length.

3.1 ^{202}Pb - ^{202}Tl a pair to determine the neutrino's upper mass limit?

The following information is extracted in context from the published article, which was generated as part of this thesis in [Wel17b].

One of nowadays ambitious ventures in physics is still the determination of the neutrino mass, which is small but not zero [Fuk98; Ahm02]. The mass determination will have a strong impact on the understanding of the origin of the fermion masses beyond the Higgs mechanism. The current upper mass limits have been determined to about 2 eV at 95% C.L.¹ for the electron-antineutrino $\bar{\nu}_e$ in tritium-decay experiments [OW08] and 225 eV 95% C.L. for the electron-neutrino mass ν_e obtained from the inner bremsstrahlung spectra of ^{163}Ho [Yas86; SBB87]. Considering a test of the CPT-violation principle [KR11], the mass of the neutrino and the antineutrino has to be determined independently with a similar sensitivity. One attempt to achieve a determination of a new upper neutrino mass limit, is the measurement of the electron capture's (EC) released energy Q_{EC} -value, which started in the 1980s at ISOLDE [Kug00]. At the time the M-shell X-rays and the Auger electrons revealed an upper limit to be $Q_{\nu_e} = 1.3(1) \text{ keV}$ for the EC in ^{163}Ho [And82], the current more precisely upper limit was determined to be $Q_{\nu_e} = 0.79(3) \text{ keV}$ with a high-precision Penning trap mass spectrometer (PTMS) [Eli15] in spectacular agreement with Cryogenic Micro-Calorimetry (CMC) [Gat97; Gas]. To reach the sub-eV level of the neutrino mass determination the results of PTMS [BNW10; BDN13] and the CMC [Ran12] a second measurement technique to obtain the Q_{EC} -value have to be combined, as the decay energy value determined by PTMS influences the atomic de-excitation spectrum following the electron capture [Klu07]. The total available energy Q_{EC} -value which is determined with a PTMS measurement, can be split into the involved reactants. In case of the EC the daughters electron capture binding energy E_{B_x} from the x^{th} atomic shell and the remaining kinetic energies of the recoil partners, including the neutrinos rest mass and its kinetic energy.

By use of a simplified EC model proposed by [Rúj82] to:

$$Q_{\nu_e} = Q_{EC} - E_{B_x}, \quad (3.1)$$

the kinetic energy and the rest mass of the neutrino can be determined to be Q_{ν_e} .

¹C.L. is the acronym for confidence level.

The simplified model, does not consider accompanying effects, among others, the electron shake-up and shake-off effects [Fae15; DRL16]. Thus, a search for other candidates to determine the neutrino mass, gains significant attention. To be a good candidate a small Q_{EC} -decay energy is of importance, which also features a small neutrino total energy and thus a high sensitivity to the neutrino rest mass at the high-energy part of the calorimetric spectrum. The criteria for choosing such alternative nuclei have been discussed in [Eli10].

In this context the electron capture pair's ^{202}Pb - ^{202}Tl Q_{EC} -value was studied more precisely at ISOLTRAP, with its large uncertainty $Q_{EC} = 46$ (14) keV [Wan12], which gave hope to consider it as a good candidate. In 2008 the ^{202}Pb -ions were produced by a hot plasma ion source [Hag92] to measure the mass more accurate. Subsequently, in 2011, an attempt was made to determine the Q_{EC} value simultaneously between ^{202}Pb and ^{202}Tl . Both species were in this case produced from a UC_x target, and ^{202}Pb was laser ionized. The measurement was, however, hampered by the very low surface-ionization yield of $^{202}\text{Tl}^+$ and by the contamination of the $^{202}\text{Pb}^+$ ground-state by its long-lived 9^- isomeric state ($T_{1/2} = 3.54$ h [Aud17]). Therefore, it was in the end necessary to determine the ^{202}Tl mass independently in a third measurement campaign in 2015, with an increased ionization yield by help of the RILIS lasers [Rot16]. This separate measurements added to the statistical uncertainties a number of systematic uncertainties of the ISOLTRAP setup. The systematic uncertainty originates from two sources. The first is a mass-dependent error, which is due to the misalignment between the electric and magnetic symmetry axes of the Penning trap. The second error has no clear mass dependence and is due to the fact that ions of different masses probe different volumes of the Penning trap and hence their frequency ratios are affected by the trap inhomogeneities (Sections 2.2.3 and 4.4.3). The statistical uncertainty of the determined mass-excess values, which are later presented, also includes the contribution of the uncertainty of the reference mass. In case of the uncertainties and how they can be qualitatively determined, the reader is referred to [Kel03] for further details.

3.1.1 Measurement

In the following the measurement of ^{202}Tl will be discussed, which was measured by use of ISOLTRAP's high-precision Penning trap. To determine the mass' cyclotron frequency ν_c , the standard well-established ToF-ICR technique by use of the single-pulse and Ramsey-type excitation schemes with various excitation times of up to $T_{ex} = 3$ s were used (Section 2.2.7). The magnetic field strength was calibrated by measuring the cyclotron frequency ν_{ref} of a reference ion with well-known mass, here, $^{181}\text{Ta}^{16}\text{O}$ and ^{203}Tl . In total 10 ToF-ICR measurements of $^{202}\text{Tl}^+$ were performed, the first six being sandwiched between ToF-ICR measurements of $^{203}\text{Tl}^+$, the last four between measurements of $^{181}\text{Ta}^{16}\text{O}^+$. The choice of using two reference ions for the measurement series ($^{203}\text{Tl}^+$ and $^{181}\text{Ta}^{16}\text{O}^+$) was made in order to avoid any bias of the determined Q_{EC} -value, due to a potential error of the literature value of

the reference mass, which was obtained during the measurement campaign for both reference masses. Nevertheless, all 10 ToF-ICR measurements of $^{202}\text{Tl}^+$ were also sandwiched between ToF-ICR spectra of $^{133}\text{Cs}^+$, in order to monitor whether there were any significant changes in the measurement conditions which would appear as sudden jumps in the frequency ratio with respect to $^{133}\text{Cs}^+$. The measured cyclotron-frequency ratios of $^{202}\text{Tl}^+$ with respect to $^{133}\text{Cs}^+$ are plotted in Fig. 3.1 (b).

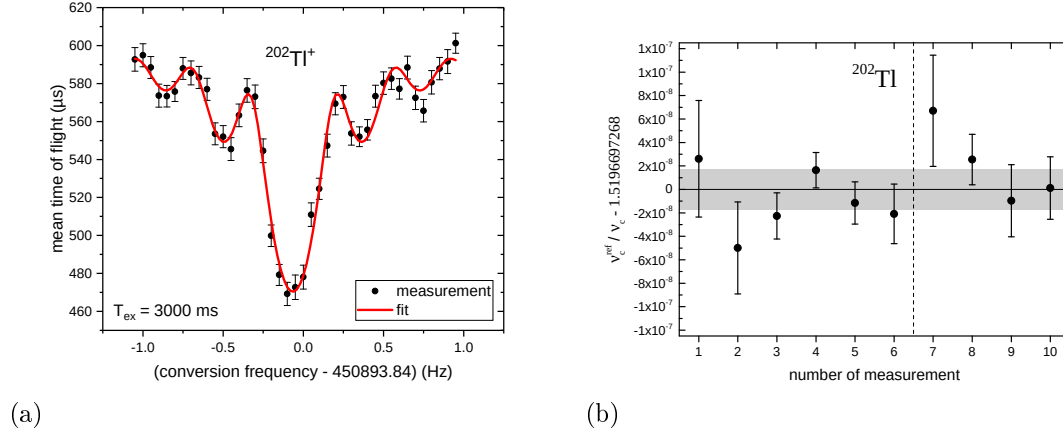


Figure 3.1: Shown is a ToF-ICR measurement of $^{202}\text{Tl}^+$ with an relatively long excitation of $T_{ex} = 3000$ ms in (a), whereby the lowering of the data points around the center resonance can be described by interaction with residual buffer-gas and the associated reduction of the cyclotron radius and the enhancement of the magnetron radius, as discussed in Sections 2.2.5 and 4.4.4. In (b) the measured cyclotron-frequency ratios of $^{202}\text{Tl}^+$ with respect to $^{133}\text{Cs}^+$ are shown. The error bars of the individual measurements are the statistical uncertainties. The dashed line in the figure separates the six $^{203}\text{Tl}^+$ and the four $^{181}\text{Ta}^{16}\text{O}^+$ data points which were used as reference. The grey shaded band represents one standard deviation of the average cyclotron-frequency ratio. For details see text.

The final obtained cyclotron-frequency ratios $r = \nu_{c,\text{ref}}/\nu_{c,\text{IOI}}$ are listed in Table 3.1:

The Birge ratio² for ^{202}Pb was not traceable and also not provided in the publication of [Boe14]. In case of the evaluation of ^{202}Tl by use of the reference ions $^{181}\text{Ta}^{16}\text{O}^+$ and $^{203}\text{Tl}^+$, a deviation over one standard deviation is noticeable, with respect to the evaluated mass-excess value with $^{133}\text{Cs}^+$ as a reference, as shown in Table 3.1. Thus, both reference masses were determined with $^{133}\text{Cs}^+$ separately. The determined masses were sent to the atomic mass evaluation community. Therefore, the here used evaluated mass-excess value of $\text{ME}(^{202}\text{Tl}) = -25980.2$ (1.6) keV [Wan17] already consists of the newly determined masses which are considered with respect of the AME algorithm that takes all masses in the entire AME network into account, including the evaluated reference masses. It should be mentioned here that the mass excess of $^{202}\text{Tl}^+$ determined by use of the mass of $^{133}\text{Cs}^+$ shown in Table 3.1, is not included in the evaluation of the AME value. This step was taken because

²The square root of the reduced chi-square is also called the Birge ratio [Bir32].

Table 3.1: Cyclotron-frequency ratios of $^{202}\text{Pb}^+$, with respect to $^{133}\text{Cs}^+$ $\text{ME}(^{133}\text{Cs}) = -88070.931(8)$ keV/ c^2 and $^{202}\text{Tl}^+$, with respect to $^{181}\text{Ta}^{16}\text{O}^+$ $\text{ME}(^{181}\text{Ta}^{16}\text{O}) = -53178.6(18)$ keV/ c^2 , as well as $^{203}\text{Tl}^+$ $\text{ME}(^{203}\text{Tl}) = -25760.8(13)$ keV/ c^2 [Wan12]. The $^{202}\text{Pb}^+$ result is taken from [Boe14]. The mass excess deduced in this work is also shown (ME_{exp}), compared to the literature value (ME_{lit}) [Wan12]. The sixth column lists the differences between the experimental value and the AME value, while the last column shows the birge ratios ($B.r.$) of the individual measurements. The statistical uncertainties are given between parentheses, while the systematic uncertainties are given between square brackets.

Ion	Reference	$r = \nu_{\text{ref}}/\nu_c$	ME_{exp} (keV)	ME_{lit} (keV)	Δ (keV)	$B.r.$
$^{202}\text{Pb}^+$	$^{133}\text{Cs}^+$	1.5196700577(284)[164]	-25941.4 (3.5)[2.0]	-25940 (4)	1.1(5.7)	
$^{202}\text{Tl}^+$	$^{181}\text{Ta}^{16}\text{O}^+$	1.0255363752 (99)[84]	-25984.5 (2.6)[1.5]*	-25986 (14)	1.5 (14)	0.36
$^{202}\text{Tl}^+$	$^{203}\text{Tl}^+$	0.9950720535(63)[80]	-25978.9 (1.8)[1.5]*	-25986(14)	7.1 (14)	0.43
$^{202}\text{Tl}^+$	$^{133}\text{Cs}^+$	1.5196697413(72)[271]	-25980.6 (0.9)[3.4]	-25986 (14)	5.4 (14)	1.44

*The mass excess used within this section is -25980.2 (1.6) keV [Wan17].

the two masses $^{181}\text{Ta}^{16}\text{O}^+$ and $^{203}\text{Tl}^+$ were determined with ISOLTRAP's $^{133}\text{Cs}^+$ reference mass independently. If the determination of $^{202}\text{Tl}^+$ with $^{133}\text{Cs}^+$ had been taken into account, whereby the measurements would no longer be independent, a systematic error could influence the mass determination of the other two.

3.1.2 Results

By use of the introduced simple approach Eq. (3.1) to determine the energy excess Q_{ν_e} for the neutrino rest mass and its kinetic energy transfer in comparison with the obtained $Q_{EC} = \text{ME}(^{202}\text{Pb}) - \text{ME}(^{202}\text{Tl}) = 38.8$ (4.3) keV it is possible to draw the following conclusion, as shown in Table 3.2:

Table 3.2: Presented are the properties of ^{202}Pb , the spin, half-life [Aud17; Zhu08] and the electron binding energies of the daughter nuclide ^{202}Tl [Lar77]. The Q_{ν} -value is deduced from the capture energy of each shell and the the Q_{EC} -value which is taken from the current section. The capture of an electron from E_{B_K} is energetically forbidden. The numbers in brackets represent the uncertainties in the last digits.

I^π	$T_{1/2}$ (ky)	$E_{B_{K1}}$ (keV)	$E_{B_{L1}}$ (keV)	$E_{B_{M1}}$ (keV)	Q_{EC} (keV)	Q_{ν_e} (keV)
0^+	52.5 (2.8)	85.530	15.348	3.704	38.8 (4.3)	L_1 : 23.5 (4.3); M_1 : 35.1 (4.3)

The binding energy of the K -electron is higher than Q_{EC} , with $E_{B_K} = 85.530$ keV [Lar77] for ^{202}Tl , hence the K -capture is energetically forbidden and thus electron capture only from higher atomic shells for ^{202}Pb is allowed.

By us of the results obtained in Table 3.2 the lowest achievable Q_{ν_e} in the case of ^{202}Pb is for the $L1$ -capture and is equal to $Q_{\nu_e} = 23.5$ (4.3) keV. In comparison to the measured value for ^{163}Ho , which is only $Q_{\nu_e} = 0.79$ (3) keV [Eli15], the EC ^{202}Pb has a large value. Therefore, it is expected that the EC in ^{202}Pb has much less sensitivity to the neutrino rest mass than the EC in ^{163}Ho , even taking into consideration the additional effects, like the shake-off of a second electron, elaborated in [Fae15;

[DRL16](#)]. Additionally, the energy regime in ^{202}Pb with its Q_{EC} -value is relatively large for nowadays cryogenic micro-calorimeters [[Gas](#); [Cro14](#); [Alp15](#)]. Last but not least the daughter nuclide ^{202}Tl is not stable and decays with a half-life of ($T_{1/2} = 12.3$ (0.08) d) [[Aud17](#)], which produces significant background in the calorimetric spectrum.

3.1.3 Conclusion

However, holmium remains the best candidate, it was demonstrated, that ^{202}Pb is an applicable candidate for CMC measurements, if further reconstructions are considered, e.g. to handle the higher induced energy in the calorimeter. The reduction of the large remaining uncertainty of the alternative candidate pair ^{202}Pb - ^{202}Tl to determine the neutrino mass with $Q_{\text{EC}} = 46$ (14) keV was achieved with a threefold higher precision to $Q_{\text{EC}} = 38.8$ (4.3) keV. However, the Q_{EC} -value has to be determined even more precisely with the new generation of PTMS (e.g. [[Rep12](#); [Rou12](#)]), to approach the sub-eV level of the neutrino mass determination by PTMS. A remaining question that has to be clarified in a separate beam time is to exclude any mass deviation that might exist for the two reference masses ^{203}Tl and $^{181}\text{Ta}^{16}\text{O}$. For this purpose, a carbon cluster reference (e.g. C_{18}) could be used to minimize the contribution of the systematic errors.

3.2 ^{79}Cu and its relevance for the determination of the ^{78}Ni 's mass surface

Most of following text is extracted in context from the published article [Wel17a], as part of this thesis.

A strong focus of today's experimental and theoretical research at the nuclear-structure of isotopes is around the region of ^{78}Ni . It is expected that this isotope has a doubly-magic character, as it lies at the intersection of the classical proton and neutron shell closures $Z = 28$ and $N = 50$, respectively. The interest is driven by the possibility of a magicity break down as it was shown in light, exotic nuclei, with the well-documented examples of $N = 20$ and $N = 28$, as reviewed in [Sor08]. It is of importance to reveal whether the exotic ^{78}Ni retains the exceptional stability of the classic closed-shell nuclides or not. Nuclear models, which extrapolate the neutron drip-lines rely heavily on the knowledge obtained from properties of nuclides closer to stability. It should be noted that T. Otsuka [Ots05] proposed that one of the fundamental parameters in the shell model is the tensor force. With the advances in effective field theory and in-medium similarity renormalization group methods [Bog14], shell-closure signatures become important links between shell-model and *ab-initio* approaches, as recently illustrated for the purported magic numbers $N = 32$ [Wie13; Ros15; GR16] and $N = 34$ [Ste13; Ste16]. In a very recent example the structure of ^{78}Ni was described by a large-scale shell-model calculation [Now16] and a coupled-cluster calculation using a chiral Hamiltonian [Hag16]. The two calculations agree on the excitation energy of the first 2^+ state, suggesting a doubly-magic character. However, the used shell model calculation (PFSDG-U), which will be described later in detail, also predicts an intruder 0_2^+ state at lower excitation energy than the 2^+ state, hinting at a new (fifth) island of inversion following those at $N = 8, 20, 28$, and 40 [Now16].

In a first attempt to address the doubly-magic character of ^{78}Ni with one of the ground-state properties, a beta-decay half-life measurement [Xu14] crossing the nickel chain at $N = 50$, showed a sudden drop in the half-lives beyond ^{78}Ni . Further studies by laser spectroscopy, probing the magnetic moment and the charge radius of the $1/2^+$ isomer of ^{79}Zn in vicinity of ^{78}Ni offer evidence for shape coexistence³ [Orl15; Yan16]. Another perspective obtained by determining the binding energies by means of mass measurements would provide decisive information, since any nuclide's configuration minimizes the ground-state energy. Unfortunately, the direct access of ^{78}Ni for mass measurements at ISOLDE is currently denied due to the 100 times lower vapour pressure compared to copper and the higher melting point,

³Shape coexistence describes that such a nucleus has several metastable ground states with different excitation energies. This leads to the behavior that on the one hand, the lowest occupied ground state has a spherical shape, for example, because the quadrupole moment vanishes. On the other hand, a decay into a higher excited ground state can lead to a deformed nucleus (e.g. prolate or oblate), as in case of ^{186}Pb [And00].

which hampers the diffusion process from an ISOL target. Thus, the subsequent discussions within this section will reflect the copper mass surface one proton above ^{78}Ni obtained at ISOLTRAP in 2015 and its relevance to the structure of ^{78}Ni .

3.2.1 Measurement

The high-precision mass measurements of the $Z = 29$ copper isotopes $^{75-79}\text{Cu}$ presented in this work are five to 100 times more precise than previous values obtained via a deflecting magnetic dipole at the Holifield Radioactive Ion Beam Facility [Hau06]. The reaction products were extracted from the target by a sequence of diffusion and effusion processes, which were subsequently ionized in a heated tantalum cavity, as in case of the surface ionization products Rb and Ga. In all measurement cases for the MR-ToF MS the Ga contamination was used as reference. The copper isotopes of interest were ionized by the resonance-ionization laser ion source (RILIS) [Fed12]. The production yield and half-lives of the copper isotopes at ISOLDE and with an uranium-carbide target adapted with a neutron converter are shown for comparison in Fig. 3.2:

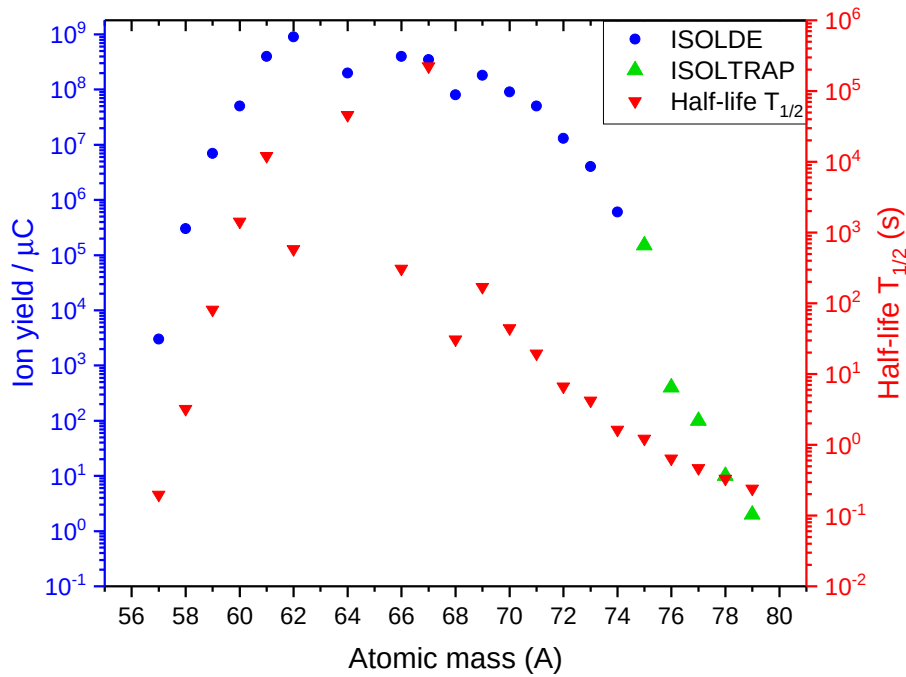


Figure 3.2: Copper yields obtained from the ISOLDE yield database [Iso], where the five last masses ($A = 75-79$) were determined during the run with ISOLTRAP’s MR-ToF MS and a total transport efficiency of 3%. The half-life information are from [Aud17].

After the ions reached the ISOLTRAP setup, they were bunched, cooled and mass separated, as discussed in Chapter 2. Only the copper ion of interest and its isobaric

impurities remained. In this case, primarily gallium and rubidium isotopes, whose yields were between 10^2 and 10^4 times more abundant than the IOI. In case of the masses $^{75-78}\text{Cu}$ the yields and half-lives were sufficient to measure the IOI by the ToF-ICR technique in the precision Penning trap, as shown in Fig. 3.3 (a) for ^{78}Cu . The yield of only 2 ions/ μC made it impossible to measure the mass of ^{79}Cu within the beam time by use of the precision Penning trap. Instead the MR-ToF MS was used to determine the mass of the $N = 50$ isotope. The high resolving power of the MR-TOF MS can be seen as an example for ^{79}Cu after $n_r = 1000$ revs in Fig. 3.3 (b):

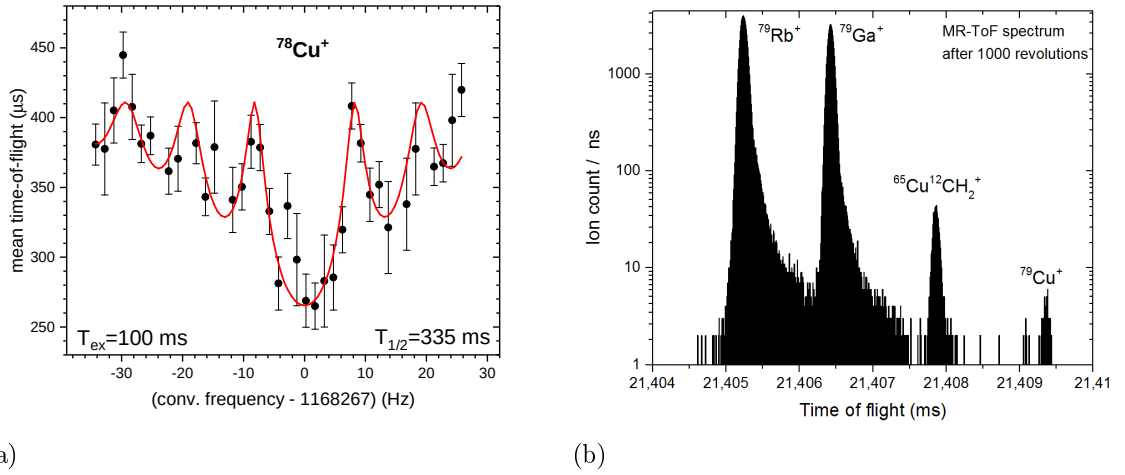


Figure 3.3: Figure (a) represents the ToF-ICR measurement of $^{78}\text{Cu}^+$, where in (b) the MR-ToF MS spectrum of $^{79}\text{Cu}^+$ and isobaric species after $n_r = 1000$ revolutions is shown. The solid line in (a) is a fit of the data to the theoretical lineshape [Kön95]. For details see text.

The contaminants in the spectrum of $A = 79$, in Fig. 3.3 (b), are $^{65}\text{Cu}^{12}\text{CH}_2$ (copper-methylene)⁴, ^{79}Rb , as well as ^{79}Ga , where the latter was used as the first reference ($m_{\text{ref}1}$) for mass determination, as described in Section 2.2.2.

3.2.2 Analysis

The MR-ToF MS was involved in all measurements of $^{75-79}\text{Cu}$ to provide purified samples for the Penning traps or direct mass measurements. The ions were trapped between 10.584 ms and 23.510 ms, which corresponds to 500 to 1000 revolutions for this mass range. For the Penning trap mass measurements, the calibration of the magnetic field $\mathbf{B}$ was performed before and after each measurement of the isotope of interest via the cyclotron-frequency $\nu_{c,\text{ref}}$ of $^{85}\text{Rb}^+$ as a well-known offline reference

⁴Copper-methylene has been seen for the first time at ISOLDE in such a production mechanism. Remarkable for this approach is that the molecule could only be produced after the copper isotope was ionized by the resonant lasers of RILIS in the hot cavity of the target and not within the target container itself. The molecule was seen on $A = 77, 79$.

isotope.

The masses of $^{75-77}\text{Cu}$ in the MR-ToF data for the to be determined masses $^{78-79}\text{Cu}$, were used to estimate the systematic uncertainty. The systematic shifts and their origin, as described below, can be demonstrated in the case of six recorded measurements of $^{75}\text{Cu}^+$ with $^{75}\text{Ga}^+$ as online reference (m_{ref1}), shown in Fig. 3.4 (a) and for twelve measurements of $^{77}\text{Cu}^+$ with $^{77}\text{Ga}^+$ as online reference (m_{ref1}), presented in Fig. 3.4 (b). For all MR-ToF measurements the second reference mass $m_{ref2} = ^{85}\text{Rb}^+$ was used, which originated from the offline alkaline source of ISOLTRAP:

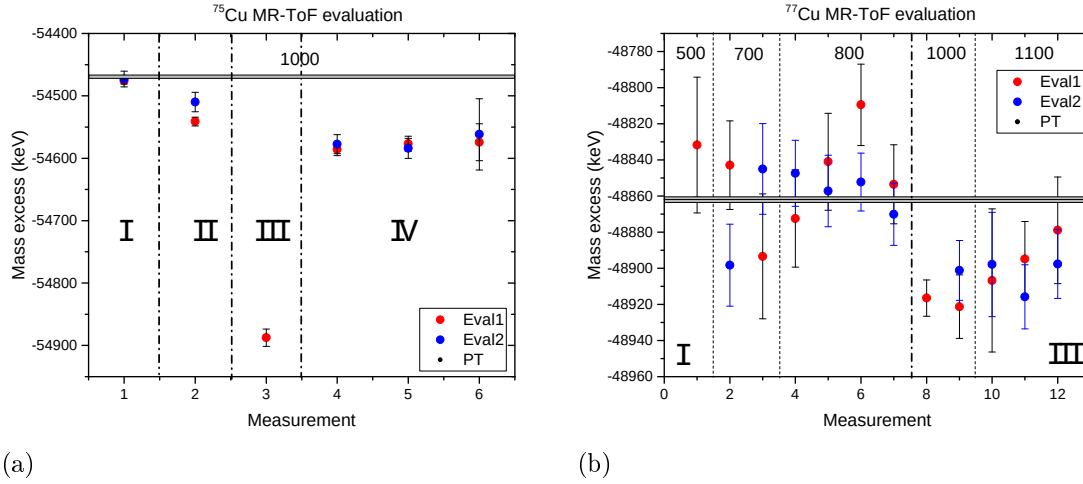


Figure 3.4: In case of figure (a), the MR-ToF mass measurements of the isotope ^{75}Cu are represented in four different sections (I-IV), each file measured at 1000 revolutions. On the right hand side (b), the measurements of ^{77}Cu are highlighted with different revolutions. The two different colors represent two different, independent data analysis obtained for each mass. The grey band represents the Penning trap measured mass and its uncertainty in both cases. For details see text.

In Fig. 3.4 (a) and (b), the situation (I) is presented as a reliable benchmark measurement in comparison to observed problems (II-IV), which can lead to incorrect flight times. These issues are among others, changing the cavity switching time between different measurement samples Fig. 3.4 (a) (II) in which the ions experience different electrostatic fields. A further problem can arise if the reference mass or the mass to be determined is in the fringe field⁵ of the cavity, as in the case of Fig. 3.4 (a) and (b) (III), in which the ions perceive a field distortion. In addition, the same effect that leads to electrostatic field turbulences occurs when the double in-trap lift (Section 2.2.2) is used to remove a contaminant during a mass measurement. Consequently, in all three aforementioned cases, the ions follow different trajectories than the reference masses, resulting in different flight times. In a last test scenario, the beam gate was kept constant during the recording of all MR-ToF measurements to

⁵Fringe fields arises at the end of electrostatic drift elements, where different potentials face each other, which in turn leads to electrostatic field distortions.

see the effect of a shift, while the proton beam now hits the target directly instead of the neutron converter. This step increased the contaminant rate of different species, therefore, more ions have reached the MR-ToF at the same time, respectively, a shift due to Coulomb interaction occurred, as can be seen in Fig. 3.4 (a) (IV).

All MR-ToF masses were evaluated with two independently developed programs. One using the Gauss fit method⁶ and another using an exponential Gaussian hybrid fit function⁷. In all cases, the masses agree within better than 1.5 sigma. The resulting mass values of each evaluation are combined in an average weighted mean. The uncertainties of the determined masses $^{78-79}\text{Cu}$ contain thereby the systematic uncertainties, which were determined from the $^{75-77}\text{Cu}$ analysis.

In the case of the ToF-ICR measurements, two different excitation patterns, the usual ToF-ICR excitation and the Ramsey-type excitation scheme were used (Section 2.2.7). The excitation timing patterns of the Ramsey-type $\tau_{rf}^{on} - \tau_{rf}^{off} - \tau_{rf}^{on}$ used in this experiment were 60-480-60 ms for ($A = 75$), 40-320-40 ms for ($A = 76$) and 20-160-20 ms for ($A = 77$), where in case of ($A = 78$) a common one-pulse ToF-ICR excitation with 100 ms was used.

The entire obtained measurement data set consists of three ToF-ICR measurements in case of ^{75}Cu , three of ^{76}Cu , four of ^{77}Cu , and one for ^{78}Cu (Fig. 3.3 (a)). For the observed MR-ToF MS-masses a total of five ^{78}Cu spectra, with revolutions two of 700 and three of 1000, as well as two measurements of ^{79}Cu (Fig. 3.3 (b)) with one of 700 and one of 1000 revolutions were recorded and contributed to the mass evaluation. In total more than 300 ions of $^{79}\text{Cu}^+$ were collected. The results of the measurement campaign are summarized in Table 3.3:

Table 3.3: Frequency ratios ($r = \nu_{c,ref}/\nu_{c,IOI}$), time-of-flight ratios (C_{ToF}) and resulting mass excesses (ME_{exp}) of the copper isotopes measured in this work. Mass-excesses values from the Atomic-Mass Evaluation 2016 (AME16) [Wan17] are also given (# indicates extrapolated values - note that the authors of the AME considered these extrapolations to be more reliable than the measured values of [Hau06]). The atomic masses of the reference masses used in the MR-ToF evaluation are $m(^{78}\text{Ga}) = 77931608.8(2) \mu\text{u}$, $m(^{79}\text{Ga}) = 78932852.3(2) \mu\text{u}$ and $m(^{85}\text{Rb}) = 84911789.738(5) \mu\text{u}$ (from [Wan17]). Experimental half-lives are taken from ^{75}Cu [Kra91], ^{76}Cu [Win90], ^{77}Cu [Ily09; Pat09] and $^{78,79}\text{Cu}$ [Gro09; Xu14]. The yield values are estimated from the approximate 3% ISOLTRAP efficiency for the MR-ToF MS. The numbers in brackets represent the uncertainties in the last digits.

A	Yield (Ions/ μC)	Half-life (ms)	Reference	$r = (\nu_{c,ref}/\nu_{c,IOI})$ or C_{ToF}	Mass Excess (keV)	
					this work	AME16
75	1.5×10^4	1224 (3)	^{85}Rb	$r = 0.8825801431$ (95)	-54470.04 (76)	-54471.3 (2.3)
76	4×10^2	637.7 (5.5)	^{85}Rb	$r = 0.8944012520$ (115)	-50981.55 (89)	-50976 (7)
77	1×10^2	467.9 (2.1)	^{85}Rb	$r = 0.9062050439$ (150)	-48862.8 (1.2)	-48520#(150#)
78	1×10^1	330.7 (2.0)	^{85}Rb	$r = 0.918033767$ (213)	-44772 (17)	-44500 (500)
79	2	241.3 (2.1)	$^{78}\text{Ga}, ^{85}\text{Rb}$	$C_{ToF} = 0.49703172$ (352)	-44819 (22)	-41740#(300#)
			$^{79}\text{Ga}, ^{85}\text{Rb}$	$C_{ToF} = 0.4963169$ (192)	-42408 (105)	

⁶Credits to the software of Frank Wienholtz and Maxime Mougeot.

⁷Credits to Marco Rosenbusch.

3.2.3 Results

The determined mass-excesses, as summarized in Table 3.3, can be used to describe the mass surface of copper until $N = 50$ with a finite-difference mass model, e.g. the two neutron separation energy (S_{2n}), as introduced in Section 1.5. In the following, the obtained experimental mass results are compared to large-scale shell-model calculation (SM) by use of the JUN45 [Hon09] and the PFSDG-U [Now16] interaction to describe the promoting behavior of protons across $Z = 28$ and neutrons across $N = 50$.

Before starting the discussion about the achieved results, the used shell-model calculation and their valence space⁸ will be introduced. As mentioned previously in Section 1.4.2, the protons and neutrons can be described as orbitals with specific occupation numbers located in a Woods-Saxon potential. This potential can be generated by blocking specific degrees of freedom, respectively, blocking levels which can be promoted by protons and neutrons. This will be used to describe the interaction space of JUN45 Fig. 3.5 (a) and PFSDG-U Fig. 3.5 (b) interaction:

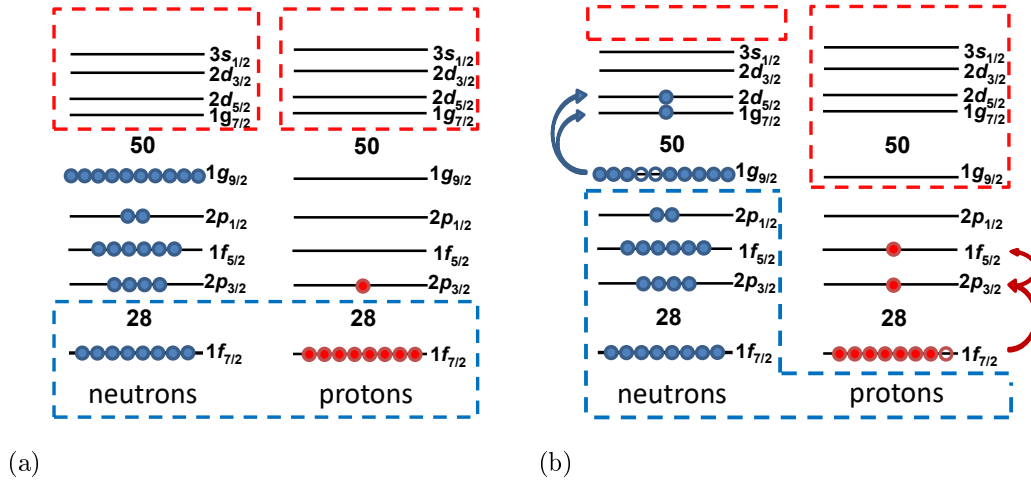


Figure 3.5: In (a) the JUN45 valence space of $N = 28 - 50$ is shown with the ^{56}Ni inert core, where in case of the PFSDG-U interaction, presented in (b), ^{60}Ca is the inert core. The inert core (blue dashed line), the external space that prohibits all the remaining orbits to be occupied (red dashed line) and the valence space in between.

The JUN45 interactions starting from a ^{56}Ni inert core⁹, for which the two-body matrix elements are optimized to describe the properties of nuclei, with protons and neutrons restricted to the $f_{5/2}, p_{3/2}, p_{1/2}, g_{9/2}$ shell between $N; Z = 28$ and $N; Z = 50$. On the opposite side (b) the PFSDG-U interaction is the model space, which incorporates the degrees of freedom required for the mass description in the range $40 \leq N \leq 52$. This interaction spans a model space comprising $f_{7/2}, p_{3/2}, f_{5/2}, p_{1/2}$ orbitals for protons and $g_{9/2}, g_{7/2}, d_{5/2}, d_{3/2}, s_{1/2}$ orbitals for neutrons and has the

⁸Valence space: orbits that contain the physical degrees of freedom relevant to a given property.

⁹Inert core: orbits that are always full.

advantage of incorporating both $g_{9/2} - g_{7/2}$ and $d_{5/2} - d_{3/2}$ neutron spin-orbit partners, as shown in Fig. 3.5 (b). The configurational valence space is extended up to eight particle-hole excitations across the $Z = 28$ and the $N = 50$ gaps and ensures convergence of the wave-functions. The exclusion or blocking of different orbitals (inert core) also blocks the possibilities of single-particle interaction between $N - N$ and $N - Z$ interaction with the core nucleons. For example, as in case of the blocked neutrons in PFSDG-U Fig. 3.5 (b), the $f_{5/2}$ neutrons are excluded from the single-particle interaction between the neutrons across $N > 36$ and protons across $Z > 20$. They are included already in the effective potential (the mean field), with which the valence particles can interact with.

As the used model (SM) spaces are now defined, the obtained calculated results can be compared with the experimental determined masses.

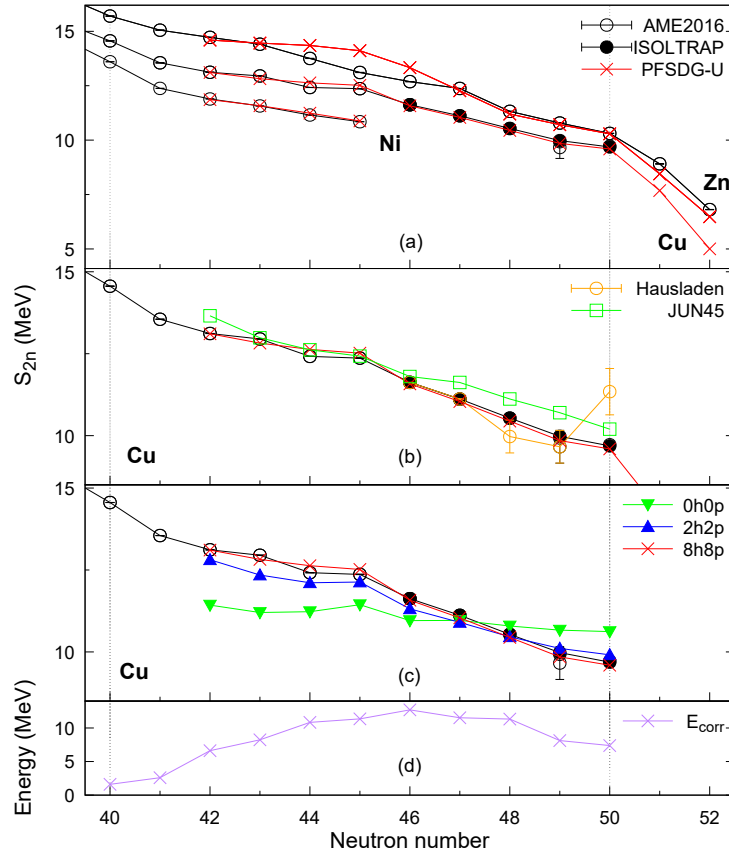


Figure 3.6: (a) Experimental two-neutron separation energies along the nickel, copper and zinc chains compared to large-scale shell-model calculations using the PFSDG-U interaction (crosses). The experimental data are from the AME2016 [Wan17] (open circles), ISOLTRAP (full black circles). (b) Comparison of S_{2n} values calculated with the PFSDG-U and JUN45 (open green squares) interactions with the present experimental data, the AME2016 and previous results [Hau06] (open orange circles). (c) Evolution of the calculated S_{2n} values in the copper chain as a function of the number of particle-hole excitations across the $Z = 28$ and $N = 50$ gaps. (d) PFSDG-U correlation energies for the $^{69-79}\text{Cu}$ isotopes.

In Fig. 3.6(a) the two-neutron separation energies $S_{2n} = -M(N, Z) + M(N-2, Z) + 2M_n$ of the nuclides in the ^{78}Ni region are shown. Here, $M(N, Z)$ represents the nuclide mass excess and M_n is the neutron mass excess. Fig. 3.6 (a) highlights the excellent agreement between the calculation and the experimental results, which gives confidence in the underlying SM-description of these exotic nuclides, with the PFSDG-U $8h8p$ -excitation, as the correct effective interaction (red crosses). The open circles in Fig. 3.6 (a) represent data from the Atomic-Mass Evaluation 2016 [Wan17]. As introduced in Section 1.5.1, a clear and sharp drop in S_{2n} at the crossing of $N = 50$ for $Z \geq 30$ (Zn), which is in most cases a well-known sign for presence of magicity [Wol13a]. It is of importance to block the correct valence space for the SM-calculation, which otherwise could generate an incorrect binding energy. This effect can be seen in case of the $N = 44 - 46$ Zn isotopes (Fig. 3.6(a)), where the uppermost PFSDG-U curve cannot correctly describe the S_{2N} trend, respectively, the nuclear mass-surface (Section 1.5). The reason for such an effect results from a suppressed mixing of the neutron orbitals between the pf and sdg valence spaces, which are known to be of increasing importance for neutron-deficient $N = 40$ isotones [Gau09]. The new trend differs significantly from the one based on measurements performed at the Holifield facility [Hau06], shown in Fig. 3.6 (b). The reliable trend of the S_{2n} values of copper based on the new measurements is linear up to $N = 49$, but abruptly changes between $N = 49$ and $N = 50$, signaling an enhancement in binding. The new masses do not extend beyond $N = 50$, therefore, it is not possible to observe the expected S_{2n} drop between $N = 50$ and $N = 52$. However, the effect of the magic number is already noticeable in the S_{2n} difference between $N = 48$ and $N = 50$ (which will be discussed at the end of the section). As can be seen in panel Fig. 3.6 (b), the S_{2n} trends differ significantly with the choice of interaction and model space. In particular, the S_{2n} values for calculations with the restricted JUN45 configuration are too strongly bound between $N = 46$ and $N = 50$ compared to the PFSDG-U result, while the S_{2n} values up to $N = 46$ are described by both interactions. The necessity of masses measured with high precision is clearly illustrated and the present results indicate the success of the new calculations over the ones with no $Z = 28$ and/or $N = 50$ core excitations. To understand the differences observed in Fig. 3.6 (b) between the Jun45 and PFSDG-U calculations, the evolution of the calculated S_{2n} as function of the number of particle-hole excitations across the $Z = 28$ and $N = 50$ shell gaps is illustrated in Fig. 3.6 (c). The asymmetry in the correlation mechanism between $N \leq 47$ and $N \geq 47$ is essentially reflecting two features: Firstly, the neutron shell closures at mid-shell $N = 40$ and closed-shell $N = 50$, and secondly, the associated particle-hole conjugation. The correlations describe an inverse parabolic behavior whose derivative should vanish at mid-shell $N = 45$ and change sign as seen in Fig. 3.6 (d). In addition, the effective single particle energies reveal a simultaneous proton and neutron gap reduction towards $Z = 28$ and $N = 50$, which slightly shifts the maximum correlation energy point to $N = 46$. The strong sensitivity of the previous correlations to ^{78}Ni -core excitations is intimately connected to the sizes of the proton and neutron gaps. From the effective single particle energies, the proton gap is considerably reduced from 6.7 MeV at

$N = 40$ to 4.9 MeV at $N = 50$. This is essentially due to the strong $f_{5/2} - g_{9/2}$ proton-neutron attractive interaction whose main active components are spin-orbit and tensor¹⁰. This also causes the $f_{5/2}$ and $p_{3/2}$ orbitals to cross between $N = 44$ and $N = 46$ as reflected in the proton orbital occupancies plotted in Fig. 3.7, which illustrates the variation of the proton orbitals with even, as well as odd-even neutron number N in the copper isotopic chain.

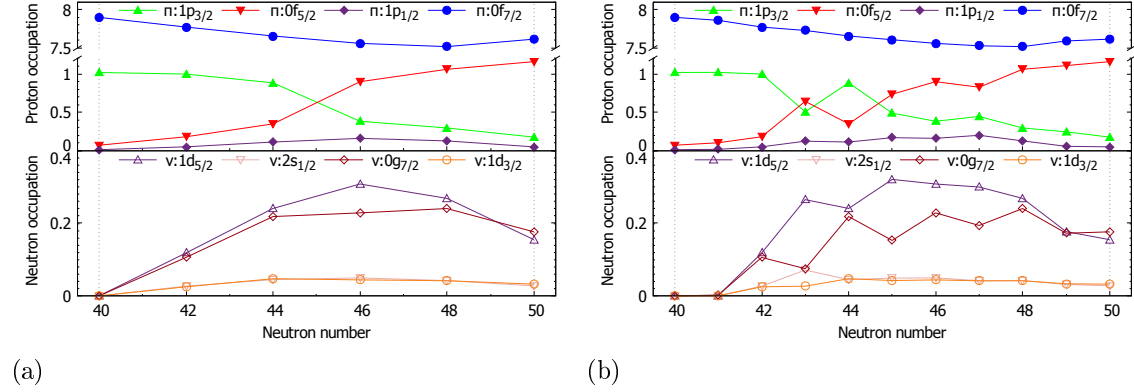


Figure 3.7: Occupation levels from the PFSDG-U calculation given in total numbers of protons and neutrons along the copper chain for even N (a), respectively, for odd and even N (b). See text for more information.

The shown promotion of the orbitals demonstrates also the slight reduction of the $f_{7/2}$ orbital occupancy as a consequence of the proton gap reduction from ^{69}Cu ($N = 40$) towards ^{79}Cu ($N = 50$). The measured $f_{5/2} - p_{3/2}$ inversion [Fla09] is perfectly accounted for by the new interaction, further re-enforcing the confidence with which the nuclear structure of this important region can be described. Recent results from laser spectroscopy [Gro17] also confirm the logic of this approach to correctly describe the g -factors of $^{76,78}\text{Cu}$. The same effect occurred in another study, when recording the β -delayed γ -ray spectroscopy of ^{77}Ni decaying to ^{77}Cu [Sah17]. Moreover, recent γ -spectroscopy of proton knockout from ^{80}Zn [Oli17] likewise point to a strong shell gap for ^{79}Cu . Interestingly to see in Fig. 3.7 (b) is the odd-even occupation behavior of the proton-neutron interaction, which causes the neutrons in $d_{5/2} - g_{7/2}$ to compensate the occupation trend of the protons. This behavior leads to a short intruder state¹¹ at $N = 43$, respectively ^{73}Cu , where $f_{5/2}$ shortly becomes the ground-state spin, before leading the configuration at $N = 46$ in ^{75}Cu onwards.

As mentioned previously, the $N = 50$ shell strength can be already probed with S_{2n} difference between $N = 48$ and $N = 50$, by use of the relation of the two-neutron shell gap (Section 1.5.1):

¹⁰The tensor force interacts with the spins of the nucleons which leads to modified single particle energies [Ots05], changes in binding energies [Pud97] and may even affect the spin-orbit splitting and the magic numbers in some cases [Ots06].

¹¹An intruder state is a swap of occupation levels that has taken the lead of the major spin assignment, in favor of lower energy states.

$$\Delta_{2n}(Z, N_0) = S_{2n}(Z, N_0) - S_{2n}(Z, N_0 + 2), \quad (3.2)$$

as a function of proton number, for the magic neutron numbers $N_0 = 28, 50, 82, 126$ in the upper curve, as shown in Fig. 3.8. The lower curve shows the shell gap calculated by use of the isotope having two neutrons *less* than the magic number: $\Delta_{2n}(Z, N_0 - 2)$. The doubly magic nuclei show a local *maximum* of $\Delta_{2n}(Z, N_0)$ at the position of the proton magic number. Without exception, for each *maximum* in $\Delta_{2n}(Z, N_0)$ there is a *minimum* in $\Delta_{2n}(Z, N_0 - 2)$ at the same proton number Z . The overall behavior of $\Delta_{2n}(Z, N_0)$ was explained in [Ben08] using a mean-field calculation, which showed that the peaked structure of the empirical shell gap is due to quadrupole correlations in the involved off-shell nuclides. The extent of quadrupole correlations in the $N_0 \pm 2$ isotopes depends strongly on Z , being *minimal* when crossing a magic number. The same argument applies for $\Delta_{2n}(Z, N_0 - 2)$. However, a local *maximum* for $\Delta_{2n}(Z, N_0)$ becomes a local *minimum* for $\Delta_{2n}(Z, N_0 - 2)$ because the binding energy of the neutron-magic nucleus appears in Eq. (3.2) with opposite sign. Observing $Z = 32$ (Ge) we note that the $N = 50$ two-neutron shell gap starts increasing towards $Z = 28$, while the $N_0 - 2$ values starts decreasing. The present data allows computing the $N_0 - 2$ value, which shows a further decrease towards $Z = 28$, as expected for a doubly magic ${}^{78}\text{Ni}$. The new copper masses thus provide evidence for the doubly magic nature of ${}^{78}\text{Ni}$.

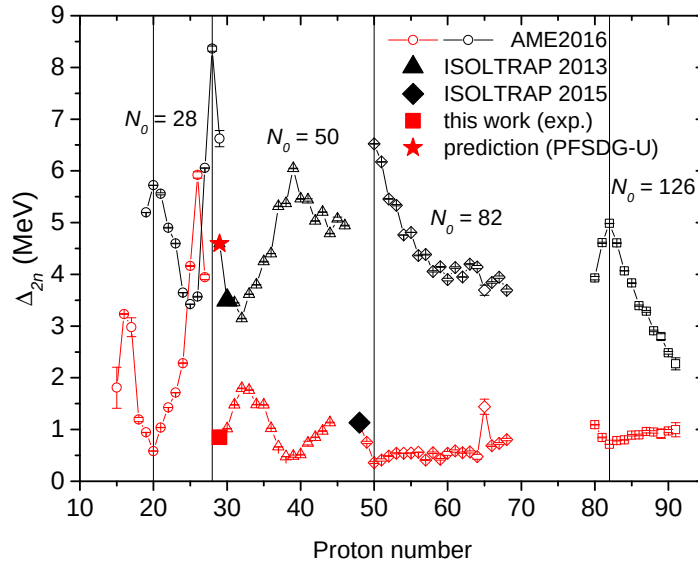


Figure 3.8: Empirical two-neutron shell gap $\Delta_{2n}(Z, N_0)$ of neutron-magic nuclides (upper curves). The lower curves represent, for each magic neutron number N_0 , the shell gap computed for $\Delta_{2n}(Z, N_0 - 2)$. The data was taken from the AME2016 [Wan17] (open symbols), [Wol13b; AW15] (full black symbols) and this work (full red symbol). See text, for details.

The new shell model results are nicely supported by the new measurements, giving

confidence in the shell-gap prediction for N_0 , which is also shown in Fig. 3.8.

3.2.4 Conclusion

The results obtained during the mass measurement studies of the neutron-rich isotopes $^{75-79}\text{Cu}$ by use of a combination of Penning-trap and time-of-flight mass spectrometry, precisely defining the mass surface above ^{78}Ni and offering evidence for its doubly magic nature. Comparison of the new experimental two-neutron separation energies to large-scale shell model calculations with the recently developed PFSDG-U interaction shows that excitations across $Z = 28$ and $N = 50$ are necessary in order to reproduce the new experimental trend, highlighting a delicate equilibrium between shape coexistence (deformed nucleus) and a spherical mean field (undeformed nucleus). The new data offer an important anchor point for calculations close to the doubly magic ^{78}Ni , strengthening their extrapolations down $N = 50$ to a possible new island of inversion [Now16]. This anchor points are important steps to describe the creation of elements in the universe, e.g. by supernova or neutron star mergers, in the r -process, as the process to produce heavy elements develops more quickly than expected [Sch05; Xu14]. Thus, the copper masses can help to understand the development of such a system more precisely.

Chapter 4

Phase-Imaging Ion-Cyclotron-Resonance technique

The at ISOLTRAP implemented phase-imaging ion-cyclotron-resonance technique (PI-ICR), as part of this thesis, was partially developed at Mainz and Heidelberg [Eit09] and later commissioned and tested at SHIPTRAP at GSI¹ in Darmstadt [Eli14]. Similar to the ToF-ICR technique, the cyclotron frequency is determined as the sum of the magnetron ν_- and the modified cyclotron ν_+ frequencies, i.e., $\nu_c = \nu_- + \nu_+$. In contrary to the ToF-ICR techniques, the cyclotron frequency ν_c by use of the PI-ICR technique is not measured by the ToF differences, but by the different phases of the radial ion motions, which are projected after a certain accumulation time in the trap onto a position-sensitive detector. The frequency is determined with the same high accuracy as ToF-ICR, but in a 25-times shorter measurement cycle and with a 35-fold gain in resolving power, compared to usual mass measurement techniques, which will be demonstrated in Section 4.2. The PI-ICR technique uses a similar effect as the former described MR-ToF MS in Section 2.2.2. In the MR-ToF MS the ions flying back and forth between a set of electrodes due to an electrostatic potential. The slight difference in mass of the trapped ions leads to a mass separation over a certain revolution time. In contrast to PI-ICR, the same effect is used, i.e., ions are stored in a weak electrostatic potential over a certain accumulation time, with the difference that this field is additionally superimposed by a strong magnetic field. Over a certain trapping time the lighter ion traveled over a greater distance, as the heavier once. This information of the ion motion in the trap is projected onto a high-resolution position-sensitive micro-channel plate (MCP) detector [Jag02], which was used in conjunction with a Penning trap for the first time in 2009 [Eit09]. The principle of the delay-line position-sensitive detector will be described in Section 4.3.1. In the following chapter the working principle of the PI-ICR technique is introduced and specifically explained, whereby the calibration of ISOLTRAP, including investigations of uncertainties, as well as a comparison between the standard ToF-ICR technique and the newly implemented PI-ICR-technique is shown in a subsequent Section 4.4.

¹GSI Helmholtz center for heavy ion research near Darmstadt.

4.1 Phase-Imaging Ion-Cyclotron-Resonance principles

In the ToF-ICR technique, the mass ratio $R_M = M_{ref}/M_{NOI} = \nu_{c_{ref}}/\nu_{c_{NOI}}$ ², is determined via the free cyclotron frequency ν_c , which was introduced in Eqs. (2.16b) and (2.56). The system response between the different conversion phases of the ions in ToF-ICR was revealed with the resulting different coupling strength and the corresponding ToF differences, as explained in Section 2.2.7. For the novel PI-ICR technique, the same principles are followed for the determination, with the difference that in the phase imaging technique ν_c is determined by measuring the magnetron Φ_- and the modified cyclotron Φ_+ motion's phase. Whereby the ions in each case evolving over an excitation-free accumulation time t_{accu} , in their specific motion. The ion's motion in the trap is disclosed in PI-ICR after the ions get ejected and by factor P enlarged projected onto the position-sensitive detector³, as shown in Fig. 4.1.

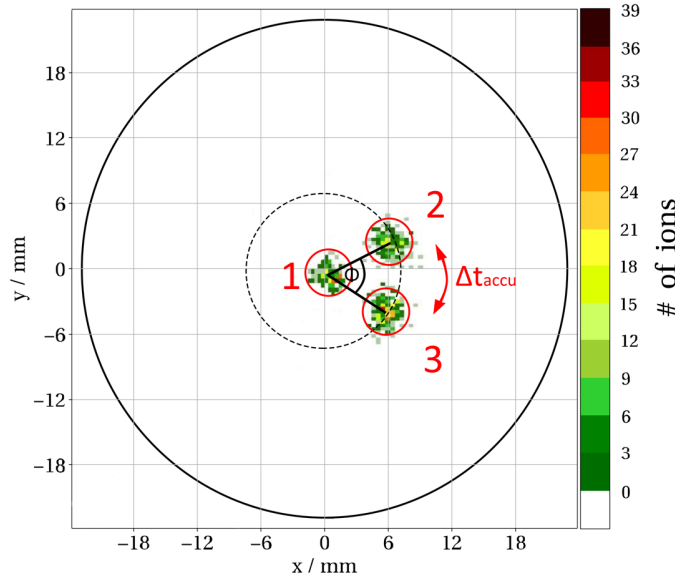


Figure 4.1: The phase-imaging projection of the ions motion in the Penning trap towards the position-sensitive detector, e.g. recorded, for $^{85}\text{Rb}^+$ ions, on a 0.5 mm magnetron radius ρ_- . The determination of the different spot positions, either in the magnetron Φ_- or the modified cyclotron motion Φ_+ , after a certain accumulation time t_{accu} are described by the rules of the inverse trigonometric functions. Highlighted are the center spot No. 1 and the ejected ion spots No. 2 and 3, on an excited radius, with the phase-difference Φ , as the to determining term to reveal the frequency of the ion species.

The spots in Fig. 4.1 are the corresponding real events and illustrate the motion sequences in a Penning trap. Spot position No. 1 defines the center of the trap,

²Here the magnetic field is omitted. $M_{ref,NOI}$, represents the reference nuclide, respectively, the nuclide-of-interest.

³The algorithm for the reconstruction is attributed to Jonas Kartheim.

with an initial spatial distribution of $2\Delta r$, corresponding to the previous cooling and correct focusing of the ions into the Penning trap. If the ions getting excited up to a certain radius $\rho_{\pm}$, they travel on that orbit at an excitation-free time t_{accu} , illustrated here in Fig. 4.1 with spot position No. 2 and at an even later time position No. 3. The phase difference between these two spots No. 2 and No. 3, referred to the anchor spot No. 1, as the center, can be determined by the cyclometric functions (inverse trigonometric functions) which allow access to the motions, respectively, frequencies.

These briefed steps are now explained in more detail using two different methods to determine ν_c with PI-ICR. I) The independent measurement scheme Section 4.1.1, where ν_- and ν_+ are determined independently and II) the direct measurement scheme of ν_c in Section 4.1.2, as mentioned in [Eli14].

4.1.1 I) Independent measurement method of ν_- and ν_+

The difference in the independent measurement scheme to measure ν_c , is almost only the dipole excitation frequency $\nu_{rf} = \nu_{\pm}$, as it was introduced in Section 2.2.4. Either the magnetron frequency ν_- or the modified cyclotron frequency ν_+ is applied, to receive $\nu_c = \nu_- + \nu_+$. The pulse-scheme for the independent measurement is shown in Fig. 4.2.

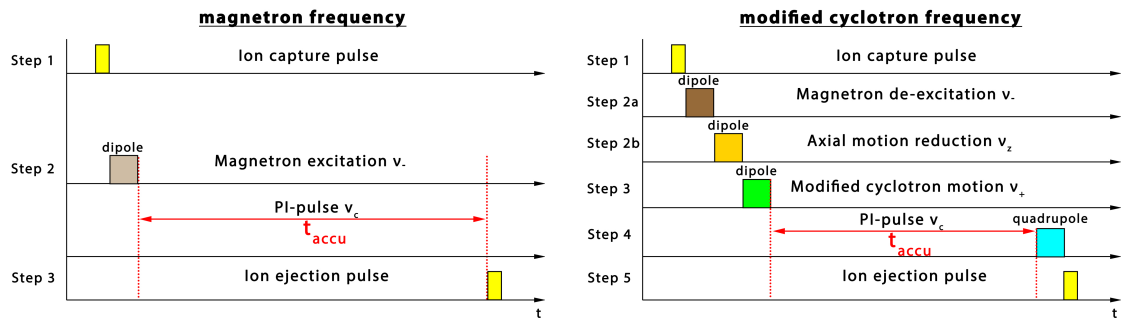


Figure 4.2: The independent measurement excitation-pulse scheme, for the magnetron (left pattern) and the reduced cyclotron (right pattern) frequencies. The excitation pulses should be as short as possible. The limitation of the pulse width is the maximum amplitude, which can be applied to convert between the motions. The so called π -pulse (right step 4), converts the reduced cyclotron motion into the pure magnetron motion. Measuring and summing over both patterns determines the free cyclotron frequency ν_c [Eli14].

The ions getting injected into the precision Penning trap, after they got cooled and re-centered in the preparation Penning trap. The center spot No. 1 in Fig. 4.1 defines the *center* in both excitation patterns. In the first measurement, the magnetron dipole excitation ν_- is applied, as illustrated in Fig. 4.2 (left). Afterwards, the ions perform a certain excitation-free accumulation time t_{accu} on the magnetron motion before they get ejected, creating the spot position No. 2 in Fig. 4.1, which highlights the *reference (ref)* position on the magnetron ring. By repeating the

measurement, the spot position would change slightly due to systematic fluctuations such as fluctuations in the magnetic field and the trap voltages. Thus, resulting in spot No. 3 the *final* (f) position. The almost same procedure is applied in a third measurement Fig. 4.2 (right), with the difference that a prior step is applied, where the magnetron dipole de-excitation pulse is used to reduce the initial magnetron motion, as explained in detail in Section 2.2.4. Without a reduction of the initial magnetron motion, the cyclotron motion would be admixed with this motion. Thus, resulting in a shifted spot distribution and with it in an additional systematic uncertainty. Subsequently, the now re-centered ions first get dipole excited at the reduced cyclotron frequency ν_+ , followed by the excitation-free evolving time t_{accu} , in the radial plane on the fast reduced cyclotron motion. After this step the ions are converted with the so called " π -pulse", a quadrupole-rf excitation at the correct sideband frequency $\nu_{rf} = \nu_- + \nu_+$, as shown in Fig. 4.2 step 4). This process performs the conversion between the reduced cyclotron motion into the pure slow magnetron motion. Then the ions getting ejected, resulting into the spot position No. 2, respectively, No. 3 in a fourth measurement, as shown in Fig. 4.1. A direct projection of the reduced cyclotron motion would be possible, but due to the axial time-of-flight distribution at ISOLTRAP of $t_{axial} = 500$ ns, as shown in Section 4.4.1, it is not suitable. This ToF difference is caused by the axial motion's energy distribution in the trap, which results into an angular spread of the spatial distribution on the position-sensitive detector, due to the fact that the ions spiral along the magnetic field lines. This effect is way less pronounced if the ions are on the slow magnetron motion, than on the fast reduced cyclotron motion. At ISOLTRAP, a typical axial ToF spread of 500 ns for ^{85}Rb results in a spatial distribution smearing corresponding to a quarter period on the detector, if the ions were ejected while they are on the reduced cyclotron motion. The simulated spatial ion distribution and its corresponding axial ion displacement is shown in Appendix A.1.

In both excitation patterns, as presented in Fig. 4.2, the ions evolve, on their radial motions in the trap over a total phase of $\Phi + 2\pi n_{\pm} = 2\pi\nu_{\pm}t_{accu}$, as shown in Fig. 4.1. The phase difference is defined by the polar angle Φ between spot position No. 2 and No. 3, where $n_{\pm}$ is the integer number of full revolutions the ions have experienced over t_{accu} , on each motion. To determine the number of full revolutions $n_{\pm}$, a conventional ToF-ICR measurement has to be performed every few days, due to moderate magnetic field drifts and small fluctuations of the trap potentials. An additional way to obtain the correct revolution number would be to extrapolate known frequencies and tune the correct frequency by use of the position-sensitive detector spot distribution shape and the ToF information. The determination of the phase difference and the corresponding evolved trajectory on the detector reveals the frequency of each motion ν_- and ν_+ independently. In independent measurement scheme, the information of the three spots is used to describe with the cyclometric functions the phase Φ of the ions' motion, respectively, the radial frequencies $\nu_{\pm}$, independently, written as:

$$\nu_{\pm} = \frac{\Phi + 2\pi n_{\pm}}{2\pi t_{accu}}, \text{ with } \Phi = 2\pi - (\text{atan2}(Y_f, X_f) - \text{atan2}(Y_{ref}, X_{ref})). \quad (4.1)$$

The angle Φ between the vectorized coordinates of the *final* phase (f) ($X_f \pm \Delta X_f, Y_f \pm \Delta Y_f$) and the *reference* phase (ref) ($X_{ref} \pm \Delta X_{ref}, Y_{ref} \pm \Delta Y_{ref}$) is described by the *atan2* function, covering 360° of the detector between $-\pi < \Phi < \pi$. To define the tangential function for all four quadrants in the polar coordinate system, a mathematical phase-shift of Φ by 2π is applied, as shown in Eq. (4.1).

4.1.2 II) Direct measurement method of ν_c

The direct measurement method allows a direct determination of the free cyclotron frequency ν_c . Compared to the independent measurement scheme described above, only two measurements, one in each pattern at a time, have to be recorded. Additionally, pattern 1 has changed, whereby pattern 2 is the same, as shown in Fig. 4.3:

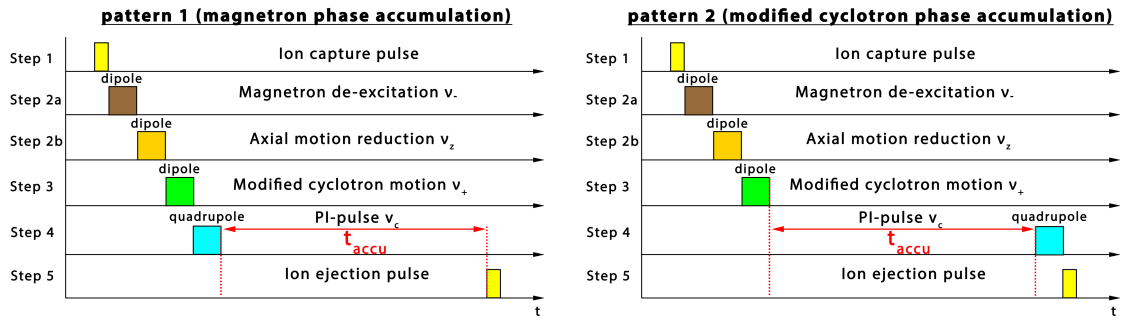


Figure 4.3: Here the direct measurement excitation-pulse scheme of ν_c is illustrated. Two different patterns are necessary to determine the phase between the two different motions. The duration of the ions in the magnetron motion is longer in pattern 1, whereas in pattern 2 the ions remain longer in the reduced cyclotron motion for a certain accumulation time t_{accu} . If for both patterns t_{accu} is equal most of the systematic uncertainties are canceling each other [Eli14]. For more information see text.

Due to the fact that an initial magnetron radius is crucial for the cyclotron motion, which would result in a mixture of both motions as shown in Fig. 4.23, a dipole de-excitation Fig. 4.3 Step 2a) at the magnetron frequency is applied, with a phase shift of $\Delta\Phi = 3\pi/2$, as explained in Section 2.2.4. Optionally, a de-excitation pulse on the end caps at ν_z can be applied to reduce the initial axial motion even further in Step 2b). This step is not yet implemented at ISOLTRAP and is just mentioned for the sake of completeness. In a subsequent step, a dipole excitation at the modified cyclotron frequency ν_+ induces energy into the radial motion Fig. 4.3 step 3). The difference between pattern 1 and pattern 2⁴ is step 4), as shown in Fig. 4.3. It is the step of converting the fast reduced cyclotron motion into the slow magnetron motion by means of a π -pulse. Thus, in pattern 1 the ions are converted immediately after

⁴Pattern 2 is equal in both methods, the independent and the direct measurement scheme.

the dipole excitation pulse at the reduced cyclotron frequency ν_+ into the slow magnetron motion, where they evolve over a certain accumulation time t_1 in the trap before they get ejected. However, in pattern 2 the ions evolve after step 3) as shown in Fig. 4.3 (right), over a certain accumulation time t_2 at the fast reduced cyclotron motion and right before they get ejected, they will be converted with the π -pulse into the slow magnetron motion. Therefore, the ions evolve either on the magnetron motion or the reduced cyclotron motion over a certain accumulation time t_1 or t_2 at the phase $\Phi_{\pm} + 2\pi n$ for pattern 1, respectively, pattern 2. If the accumulation time is equal $t_1 = t_2 = t_{accu}$, for both patterns, the angle between the *reference* phase and the *final* phase corresponds to Φ_c , given to:

$$\Phi_c = 2\pi\nu_c t_{accu} - 2\pi(n_- + n_+). \quad (4.2)$$

If the evolution numbers for the magnetron motion n_- and the reduced cyclotron motion n_+ over the evolution time t_{accu} are known, the free cyclotron motion results immediately from the phase difference between the pattern 1 spot No. 2 (*ref*) and pattern 2 spot No. 3 (*f*) as shown in Fig. 4.1.

4.2 A direct comparison of the novel PI-ICR and the standard ToF-ICR technique

Within this section, the improvement of the PI-ICR technique compared to the ToF-ICR technique is shown. First of all, the gain in resolving power $\mathfrak{R}$, followed by the achieved gain in precision, as well as the representation of the shorter measurement time, obtaining the same precision of ν_c as in the case of the ToF-ICR technique. Most of the formulas used within that section are taken from [Eli14].

4.2.1 Initial spatial distribution

As reviewed earlier for the ToF-ICR technique in Eq. (2.54), also the PI-ICR technique has a resolving power. Its mass resolution is limited by the ratio between the motion's radius r and the radial image spread Δr , as shown in Fig. 4.1. The initial spatial distribution limit $2\Delta r$ of the ions is determined, among other things, by the transfer and the steering electrodes from the preparation Penning trap into the precision Penning trap over a transport distance of about 2.5 m at ISOLTRAP. It is assumed that if the voltages of the transfer electrodes are tuned, the leading term of the initial spatial distribution $2\Delta r$ is determined by the motions cooling in the preparation Penning trap, as described in Section 2.2.5. The ions in the preparation Penning trap are subject of the temperature, which are in contact with the helium buffer-gas at room temperature $T = 293$ K. Thus, the probability density function for the energy E_{kin} of the cooled ions is given by the Boltzmann distribution [BG86; Bol90; Eli14]:

$$f(E_{kin})dE = \frac{1}{k_B T} e^{-\frac{E_{kin}}{k_B T}} dE. \quad (4.3)$$

Here the gas temperature is given by T , whereas k_B is representing the Boltzmann constant. By using the energy relation and the frequency hierarchy, as introduced in Section 2.2.3 and Eq. (2.24), the correlation between the radius of the cooled cyclotron motion ρ_+ and via the sideband coupling also the radius of the magnetron motion ρ_- , as described in Section 2.2.5, can be written with the following relation of the motions temperatures $T_+ = (\omega_+/\omega_-)T_-$ [BG86]. According to those relations the probability density function can be written as:

$$f_{\pm}(r_{\pm})dr_{\pm} \approx \frac{m\omega_{\pm}^2}{k_B T} e^{-\frac{m\omega_{\pm}^2 r_{\pm}^2}{2k_B T}} r_{\pm} dr_{\pm}. \quad (4.4)$$

Assuming the radial motions are not correlated with the buffer gas and the sideband cooling, there is a probability of $F(r, \Phi)$ locating an ion at a specific (r, Φ) position in the radial plane by the following convolution:

$$F(r, \Phi) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f_+(r_+, \Phi_+) f_-(r - r_+, \Phi - \Phi_+) dr_+ d\Phi_+. \quad (4.5)$$

By use of the substitution $\sigma = \sqrt{\frac{2k_B T}{m\omega_+^2}}$, the polar coordinates can be converted into the Cartesian coordinates to:

$$\begin{aligned} F(x, y) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f_+(x_+, y_+) f_-(x - x_+, y - y_+) dx_+ dy_+ \\ &= \frac{1}{(\pi\sigma^2)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{-\frac{x_+^2 + y_+^2 + (x - x_+)^2 + (y - y_+)^2}{\sigma^2}} dx_+ dy_+ \\ &= \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{x^2}{2\sigma^2}} \cdot \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{y^2}{2\sigma^2}} \\ &= \frac{1}{2\pi\sigma^2} e^{-\frac{r^2}{2\sigma^2}}. \end{aligned} \quad (4.6)$$

Thus, the cooled and centered ions in the preparation Penning trap are described by a Gaussian distribution with a FWHM of $2\Delta r$:

$$2\Delta r \approx \frac{2.35}{\pi\nu_+} \sqrt{\frac{k_B T}{2m}}. \quad (4.7)$$

Therefore, the distribution of $2\Delta r$, at ISOLTRAP's preparation Penning trap, is corresponding to $\sim 100\mu m$ in case of $^{133}\text{Cs}^+$ at room temperature $T = 293$ K. Whether this ion distribution can be retained as a spatial spot distribution after injection into the precision trap is part of an ongoing study that will be published elsewhere. To support this important starting condition, new, almost mass-independent voltages for ion transport between the preparation Penning trap and the precision Penning trap were found within this thesis, as shown in Appendix A.2. Additionally, SIMION simulations, which include the ion transport between the LT into the UT, have been

implemented to support further studies. The simulation environment, including the magnetic fields of both traps, as well as the electrostatic drift elements, is shown in Appendix A.3.

Under this condition, the ToF-ICR resolving power Eq. (2.55) can be compared with the PI-ICR resolving power, by taking into account the spatial distribution:

$$\frac{\Delta m}{m} = \frac{2\pi\Delta\nu_c}{2\pi\nu_c} \approx \frac{1}{\Re\sqrt{N_{ion}}} = \frac{m}{qB} \frac{1}{t_{accu}\sqrt{N_{ion}}} \frac{2\Delta r}{r}. \quad (4.8)$$

The ToF-ICR resolving power $\Re_{PI-ICR}$, as previously described in Eq. (2.55), can be extended by the spatial-distribution-to-radius ratio to:

$$\Re_{PI-ICR} \sim 1.25 \cdot \nu_c \cdot t_{accu} \cdot \frac{r}{2\Delta r}. \quad (4.9)$$

The ratio obtained at ISOLTRAP with drift electrodes at certain simulated voltages, as shown later in Section 4.3, covers a wide mass range and achieves an evenly distributed mass-to-spatial spot distribution over several mass units, as shown in Fig. 4.4.

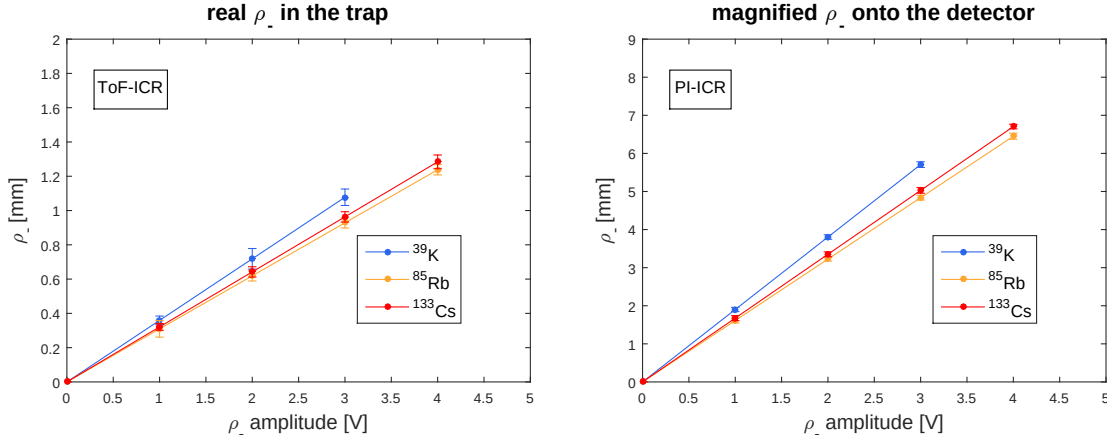


Figure 4.4: Shown are the magnetron radii, with respect to different magnetron ρ_- excitation amplitudes. On the left hand side, the picture describes the radii for the ions in the trap, determined with the ToF-ICR technique. Where on the opposite side the picture obtained by use of the PI-ICR technique illustrates the magnified magnetron radii ρ_- , which are projected onto the position-sensitive detector. The different alkaline ions available from ISOLTRAP's ion source, which are compared here, are $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$. The excitation time in both measurements lasted $T_{ex} = 1200$ ms. Thus, the resulting error bars for the PI-ICR plot are as small as the markers.

With the obtained values of Fig. 4.4 the magnification factor of the magnetron radius can be determined on average to $r_{ToF}/r_{PI-ICR} = P = 6.5$. This magnification is illustrated for ISOLTRAP's alkaline ions $^{39}\text{K}^+$, $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$ in Fig. 4.4, as a

direct comparison between the measured ToF-ICR magnetron radii ρ_- in the trap and the obtained PI-ICR magnetron radii ρ_- projected onto the position-sensitive detector. The comparison between the real observed radii magnification factor and the one simulated in the SIMION⁵ environment, agree with each other, as shown in [Kar17]. Further details for the magnification factor P will be discussed in detail in Section 4.4.6.

The already mentioned higher accuracy achievable with the Ramsey-type excitation $\delta\nu_c \sim 0.6/T_{ex}$, compared to the one-pulse scheme $\delta\nu_c \sim 0.8/T_{ex}$, was announced in Section 2.2.7. Due to the reduced possibility to resolve isomeric states, caused by superposition of the multiple fringes in the Ramsey scheme, the ToF-ICR one-pulse scheme and its values are used for further comparison. The ToF-ICR precision can now be combined with the resolving power of the projected spot on the position-sensitive detector, as introduced in Eq. (4.8) to:

$$\delta\nu_c \approx \frac{0.8}{t_{accu}\sqrt{N_{ion}}} \cdot \frac{2\Delta r}{r_-}. \quad (4.10)$$

Here, the new technique reveals its strength in the comparison between the standard ToF-ICR and the novel PI-ICR technique. The mentioned Fourier limit in the ToF-ICR technique, which defines the limits of $\Delta\nu_c$ or the resolving power $\mathfrak{R}$, as shown in Section 2.2.7, is no longer the limiting factor in case of the PI-ICR technique.

4.2.2 The resolving power in comparison

To compare which ion motion in PI-ICR contributes the most to resolve two ion species, either the slow magnetron motion or the reduced cyclotron motion, the following ansatz can be used. The representation of the resolution $\Delta\nu$ and the uncertainty $\delta\nu$ by use of the independent measurement scheme, declared in Section 4.1.1 can be written as [Eli14]:

$$\Delta\nu = \frac{\Delta\Phi}{2\pi t_{accu}} = \frac{2\arcsin\left(\frac{\Delta r_{det}}{r_{det}}\right)}{2\pi t_{accu}} \approx \frac{1}{\pi t_{accu}} \cdot \frac{\Delta r_{det}}{r_{det}}, \quad (4.11)$$

$$\delta\nu = \frac{\sqrt{\sum_{ref,f} [(Y\delta X)^2 + (X\delta Y)^2]}}{2\pi r_{det}^2 t_{accu}}. \quad (4.12)$$

Under normal conditions the equation Eq. (4.12) is reduced to Eq. (4.13), in case of an isotropic distributed spatial spread, e.g. $\delta X_f \approx \delta Y_f = \delta A$:

$$\delta\nu_{\pm} = \frac{\delta A_{\pm}}{2\pi r_{det} t_{\pm}}. \quad (4.13)$$

⁵An ion simulation program for electric and magnetic fields [Dah00].

Hence, the resolving power of the magnetron motion is dominated by the cyclotron motion. The following assumption can therefore be assumed to:

$$\frac{\nu_c}{\Delta\nu_c} \approx \frac{\nu_+}{\Delta\nu_+} = \frac{\Phi + 2\pi n_+}{\Delta\Phi} \approx \pi\nu_+ t_{accu} \cdot \frac{r_{det+}}{\Delta r_{det+}} = \pi\nu_+ t_{accu} \cdot \frac{r_+}{\Delta r_+}. \quad (4.14)$$

By use of Eq. (2.55) and Eq. (4.12), the enhancement in resolving power of the PI-ICR technique compared to the standard one-pulse ToF-ICR scheme can be estimated to:

$$\mathfrak{R}_{gain} = \frac{\mathfrak{R}_{PI-ICR}}{\mathfrak{R}_{ToF-ICR}} \approx 0.8\pi \cdot \frac{r_+}{\Delta r_+}. \quad (4.15)$$

With ISOLTRAP's typical final radius of $r_+ = 0.6$ mm, as well as the radial spread of $\Delta r_+ = 50$ μ m calculated for $^{133}\text{Cs}^+$ with Eq. (4.7), the improvement is about $\mathfrak{R}_{gain} = 35$. As a result of the above-mentioned investigations Eq. (4.14), both the center and the reference coordinates do not have to be checked so often. This reduces the independent measurement cycle to the second pattern Fig. 4.2 (right), and thus to define only the final phase's coordinates.

4.2.3 The precision of both techniques

With the sum of the uncertainties of ν_- and ν_+ , the uncertainty of the free cyclotron motion can be determined by $\delta\nu_c = \sqrt{\delta\nu_+^2 + \delta\nu_-^2}$. Assuming the ideal case, where the following conditions are given, $r_{det-} = r_{det+} = r_{det}$, $t_- = t_+ = t$, as well as $N_- \simeq N_+ \simeq N_{ion}/2$. Here, N_- determines the amount of ions recorded for the magnetron frequency measurement, respectively, N_+ for the reduced cyclotron frequency. N_{ion} indicates the total number of recorded ion events. On this assumption the uncertainties $\delta\nu_-$ and $\delta\nu_+$ are similar and can be merged by us of Eq. (4.13) to:

$$\delta\nu_c = \sqrt{2} \frac{\delta A_c}{2\pi r_{det} t_{accu}} = \frac{1}{\pi t_{accu} \sqrt{N_{ion}}} \cdot \frac{\Delta r_{det}}{r_{det}} = \frac{1}{\pi t_{accu} \sqrt{N_{ion}}} \cdot \frac{\Delta r}{r}, \quad (4.16)$$

where δA describes the uncertainty of the final magnetron or cyclotron phase. Thus, dividing Eq. (4.10) by Eq. (4.16) leads to the following precision increase:

$$\frac{(\delta\nu_c)_{ToF-ICR}}{(\delta\nu_c)_{PI-ICR}} \approx 1.6\pi \cong 5. \quad (4.17)$$

The accuracy obtained is increased by a factor of five and can also be expressed in terms of a measurement duration, since the precision Eq. (4.16) is inversely proportional to the square root of the detected ions $\delta\nu_c \sim 1/\sqrt{N_{ion}}$. Thus, Eq. (4.17) implies that the PI-ICR technique, is able to achieve the same precision 25-times faster than the standard ToF-ICR technique.

Within this section the standard ToF-ICR technique was compared to the PI-ICR technique at ISOLTRAP. Thereby it was shown that the new technique offers the ability of a 25-times shorter measurement cycle, which leads to the same uncertainty as with ToF-ICR. This allows to study regions with even shorter lived isotopes by an order of magnitude of a half-life. To be able to resolve isomeric and ground states, the PI-ICR technique offers a mass resolving power, which is on average improved by a factor $\mathfrak{R}_{gain} = 35$, compared to the one-pulse ToF-ICR technique. All discussed points for the independent measurement scheme (Section 4.1.1) are also valuable for the direct measurement scheme (Section 4.1.2). To conclude, the following illustration Fig. 4.5 visualizes the mass resolving power compared between the different techniques at ISOLTRAP, including the MR-ToF MS Eq. (2.4), the one-pulse ToF-ICR Eq. (2.55) and PI-ICR technique Eq. (4.9), as previously described in this chapter.

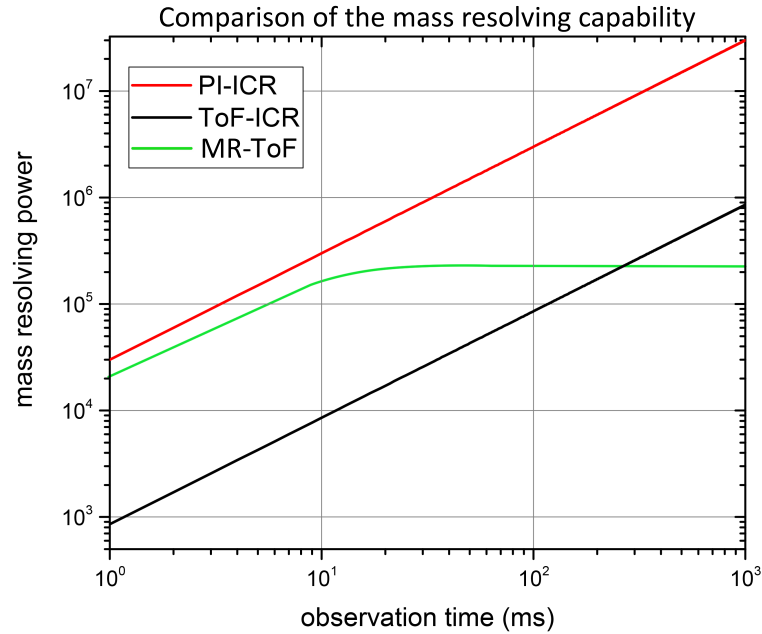


Figure 4.5: The theoretically attainable mass resolving powers of different techniques used at ISOLTRAP, as a function of the observation time. Highlighted are the MR-ToF (green), the standard one-pulse ToF-ICR (black), as well as the PI-ICR (red) technique for a mass $A = 100$ nuclide.

The resolving power limits caused by e.g. ion interaction with rest gas, as well as magnetic-field and voltage drifts in the ToF-ICR and PI-ICR technique are not visible here, due to the short observation times. Whereas in case of the MR-ToF MS the time-of-flight spread of the ion bunch and the trajectory stability are already limiting the resolving power at an observation time of $t_{obs} \approx 160$ ms⁶.

⁶Credits for the MR-ToF curve to F. Wienholtz and R. N. Wolf.

4.3 Adjustment of ISOLTRAP for the PI-ICR technique

Compared to the ToF-ICR ion ejection scheme from the precision Penning trap downstream to the detector, the PI-ICR technique requires a different ejection and transport voltage scheme. Thus, the prior ISOLTRAP ejection scheme has to be adapted to master both techniques at the same time. These adaptations are part of the following chapter. One of the biggest changes is the replacement of the standard multi-channel plate detector (MCP) by a position-sensitive delay-line detector (PS-MCP), as already mentioned previously. This will allow to preserve the necessary position and timing information for the PI-ICR technique, as described in Chapter 4. The function principle of the PS-MCP will be clarified subsequently prior the new ejection schemes.

4.3.1 The delay-line position-sensitive detector

To access the field of the phase-imaging technique a delay-line detector (DLD40) from RoentDek GmbH [Jag02] was implemented at ISOLTRAP within this work. The first studies of the magnetron motion in combination with a PS-MCP were achieved for the first time in Mainz [Eit09], before intensive studies of PI-ICR were performed by the SHIPTRAP group at GSI in Darmstadt [Eli14]. Before the working principle is described in detail, a short overview of the most important facts of the DLD40 MCP is given in Table 4.1:

active diameter	42 mm
open area ratio	> 50%
MCP channel diameter	25 μm
position resolution	70 μm
time resolution	~ 10 ns
event rate capability	< $2 \cdot 10^6$ ions/s
max. ambient magnetic field	a few 10 mT

Table 4.1: Specifications of the delay-line anode detector DLD40 from RoentDek GmbH.

The MCP of the DLD40 anode detector is a usual Ni-Cr coated MCP with two plates oriented in the chevron structure, to allow the production of an electron avalanche of about $10^6 e^-/\text{ion}$ [Wiz79]. The electron cloud arises after an ion hits the MCP front plate, which has an attractive potential of $\text{MCP}_{\text{Anode}} = -2.15$ kV at ISOLTRAP. In this voltage regime the dark count rate (N_{dark})⁷ is at an acceptable level, as well as the incident ions have enough energy to create a signal with a sufficient pulse-width

⁷The MCP dark counts have four main origins [LCN13]: first the electric field emission from the channel walls, second the ionization of residual gases, third local discharges by high electric fields and last but not least due to photo-electron emission generated by photons which are produced by electric field scintillation of the MCP support structure.

for further signal discrimination. The dark counts are within the order of ~ 120 Hz, which gives an acceptable signal-to-noise ratio at ISOLTRAP of $N_{dark} = 0.2$ ions/h, within the ion arrival ToF region $t_{axial} = 500$ ns. This count rate can be suppressed even further with the position information in a post-analysis. The physical limitations given by the time and position resolution, as presented in Table 4.1, already set the minimal *bin*-width and thus the framework conditions for further data acquisition (DAQ). These limits have to be considered for the smallest possible spot size as well as the ToF distribution with which the ions reach the detector t_{axial} . The at ISOLTRAP used delay-lines are two independent copper wire-grids, which span the x - and y -axis. Each wire represents an axis, where they are wrapped around in a consecutive manner, with a distance of $\Delta x = 70 \mu m$. The produced electron cloud is traveling towards the positively charged delay-lines with a potential of $U_{DL} = +300$ V. At its point of impact, this cloud generates a signal which propagates to both ends of each wire simultaneously, as shown in Fig. 4.6:

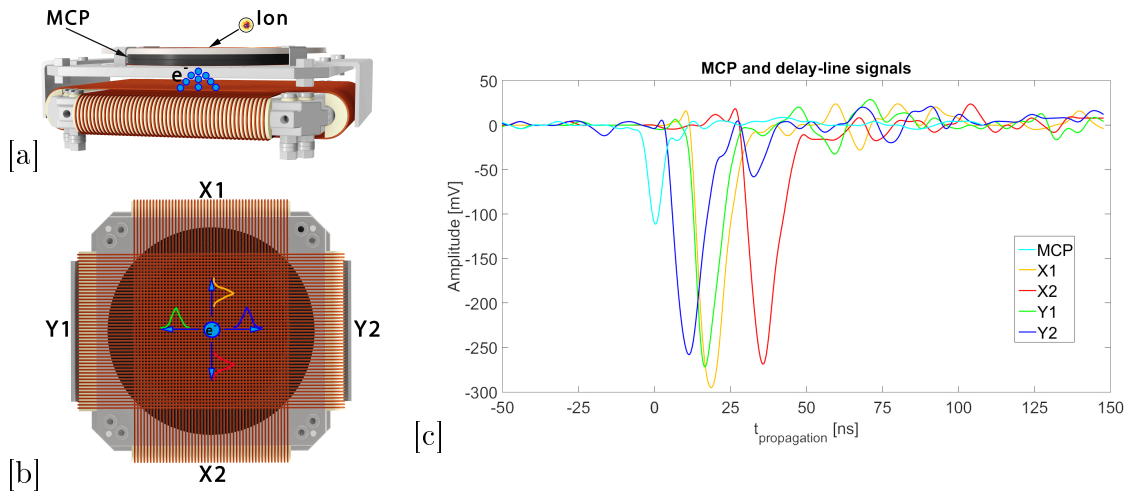


Figure 4.6: On the left hand side the position-sensitive multi-channel plate detector (PS-MCP) with its delay-lines is illustrated in a rendered CAD-drawing. The impact of an ion is implied generating an electron cloud after impinging on the MCP (a). The impact of the ion on the MCP is used as a reference timing window. The electron cloud attracted by the positively charged delay-lines creates a signal within the wire that propagates into both directions on each of the two delay-lines, as shown in (b). Visualized with a real event oscilloscope picture on the right hand side (c). With respect to the MCP acceptance window (cyan) and the two signals for the X1-X2-wire (yellow, red), respectively, the Y1-Y2-wire (green, blue) of the delay-lines can be used to locate the ions impact origin, with a minimum spatial resolution of $\Delta x = 70 \mu m$.

The difference in signal propagation time unveils the true ion impact location with respect to the x - and y -coordinates, which will be explained in more detail in Section 4.3.2. The MCP has a large active area with a diameter of about 42 mm. Thus, a large magnification of the ions motion onto the detector is possible, which is an advantageous feature for the PI-ICR precision as described in Eq. (4.9). Another effect to be taken into account is that the DLD40 does not operate in vicinity of a

magnetic field stronger than a few 10 mT. The strong magnetic field would otherwise perturb the electron clouds propagation path on their way to the delay-lines, resulting into a displacement of the true ion impact point. Thus, the detector aims to be further out with respect to the trap center, which enhances the magnification of the ions motion projection. These restrictions defining the magnification factor Section 4.4.6, as well as the layout of ISOLTRAP's electrostatic ion optics, as described in Section 4.3.3.

To emphasize the delay line detection technique for ISOLTRAP's specific purpose, different detection concepts, supported by an MCP, are summarized and compared with each other as in the following Table 4.2:

Criteria	crossed DLD	hexagonal DLD	RAE	hybrid active PD	phosphor & CCD
Detector	2 orthogonal wires	three wires at 120° to one another	four resistive anodes	256×256 pixels on chip (TP/MP)	Phosphor screen 1000×1000 pixel on CCD chip
No. of MCPs	2-3	2-3	2	1-2	2
detector size	40-120 mm	40-120 mm	25 mm	28×28 mm	40-70 mm
No. of amplifiers	5	7	5	65k on chip	2
spatial resolution	~ 17-50 μm	13 μm	1-2 μm	55 μm (MP) 5 μm (TP)	50-200 μm
time resolution	~ 130 ps - 25 ns	25 ps	1 ns	10 ns	100 ns - 33 ms
Max. count rate	1·10 ⁶ Hz	2·10 ⁶ Hz	6·10 ⁵ Hz	1·10 ⁹ Hz (MP) & 1·10 ⁶ Hz (TP)	1·10 ⁶ Hz
Detector noise	120-1000 Hz	120-1000 Hz	60 Hz	electronic noise	23 e ⁻ /pixel-s
achievable vacuum	1·10 ⁻¹⁰ mbar	1·10 ⁻¹⁰ mbar	1·10 ⁻¹⁰ mbar	1·10 ⁻⁷ mbar	1·10 ⁻⁹ mbar

Table 4.2: Given is an overview of state-of-the-art position-sensitive detector concepts for efficient ion-detection, for further details see [JH13]. To convert the incident ion into electron avalanches, a MCP as a converter is combined with different electron detection techniques. Among others, a phosphor screen and a CCD camera (CCD), resistive or cross strip anode encoders (RAE), crossed or hexagonal delay-line detectors (DLD), as well as pixel detectors (PD), either in Timepix (TP) or Medipix (MP) mode.

The hexagonal delay-line detector with a diameter of 40 mm is a new version of its size and was not available during the installation time at ISOLTRAP. The new detector would have allowed higher ion rates, due to a third wire grid, which can be used as a coincident signal trigger layer. The high count rates could be compensated with a phosphor screen technique, but the limiting factors are the luminescence decay times of up to 30 μs with the newest materials, as well as the CCD frame rate of roughly 2·10³-1·10⁴ frames/s to probe the intensity fluctuations [JH13]. It should be mentioned that the ToF information of the incident ion for all introduced solutions

in combination with an MCP is preserved and can be used for further analysis.

4.3.2 ISOLTRAP's PI-ICR data-acquisition system

The ability to achieve the position-sensitive information in addition to the ToF signal of the MCP acquires a new read-out system at ISOLTRAP. After several tests, RoentDek's equipment was chosen for the entire readout chain. The DLD40 delay-line detector, explained in Section 4.3.1, is connected to a galvanic decoupling box (FT12-TP). This allows the signals to be galvanically decoupled from the high DC (direct current) voltages of the MCP anode and the delay lines. These small signals of about ~ 2 mV are amplified by the FAMP3b⁸, with a gain of factor > 50 , to values of about > 100 mV. This sufficient signal height can be converted into the NIM-standard⁹ signals by use of a high precision constant-fraction discriminator (CFD). Since the time information of such a position-sensitive detector is a decisive factor, no leading edge discriminator can be used. A constant fraction discriminator has its trigger-threshold at an equal fraction of the signal height, therefore, the timings are more reliable than in the case of an leading edge discriminator. At ISOLTRAP the CFD discriminator leads to a small timing jitter of about $t_{jit} = 250$ ps, which causes an equivalent position jitter of $\Delta x = 0.3$ mm¹⁰. The constant-fraction discriminator at ISOLTRAP supports a NIM-signal after the conversion with a width of about ~ 10 ns. All five signals, the MCP timing information and the four delay-line signals are transmitted to a time-to-digital converter (TDC8HP). This TDC converter saves NIM signals of up to eight inputs in parallel with a time resolution of 25 ps. The internal storage capacity of this card is able to store $5 \cdot 10^5$ ions/s within a time window of $418 \mu\text{s}$ for ~ 2 h. ISOLTRAP has revised¹¹ the LabView code of the MPIK Heidelberg [Gon14], which is responsible for retrieving the time stamps from the TDC card. Thus, the stored timing information within the raw data of the TDC card can be stored in a text file for further data analysis. The software that reads the text file correlates the MCP timing trigger with the correct delay-line signals by use of a sorting algorithm, as described in [Kar17]. The algorithm is able to reconstruct the position of the ion impact, in the radial plane but can be used as well to probe the ToF distribution with which the ions reach the detector, as described in Section 4.4.1. It should be mentioned that this algorithm is capable to recreate lost signal pairs, even when signals are lost in both dimensions that might be suppressed by the CFD threshold. This ability allows a higher reconstruction efficiency in the post-analysis of up to 25% [Kar17].

⁸The RoentDek FAMP3b hosts 3 channels of bipolar amplifiers with a maximum bandwidth of 200 MHz [Roe17a].

⁹NIM is the acronym for Nuclear Instrumentation Modul-Standard.

¹⁰By use of the signal pitch propagation time within the delay-lines, recommended by RoentDek with 1.33 mm/ns [Roe17b].

¹¹Credits to Jonas Karthein and Dinko Atanasov.

4.3.3 A new ejection scheme & further changes for ISOLTRAP

Until 2014, ISOLTRAP used a slow extraction potential lift scheme, followed by an extraction pulse. For that purpose the ring electrode, which is at $U_{trap} = -10.1$ V, got lifted closer to the potential of the correction electrodes, before the extraction pulse was acknowledged. In order to be able to use the new fast PI-ICR ejection scheme and still be compatible with the ToF-ICR method, the extraction scheme had to be adapted at ISOLTRAP, as shown in Table 4.3.

Trap & drift electrodes	previous ToF-ICR [V]	new ToF-ICR [V]	PI-ICR [V]
U_{trap}/Ring	-10.1	-10.1	-10.1
Correction ring	-6.5	-8	-8
End cap	-1.62	-1.62	-1.62
Correction tube	1.5	1.5	1.5
Drift1	-11.4	-60	-150
Drift2	-2.7	-15	-150
Drift3	-2.7	-14	-320
Drift4	-42	-15	-320
Drift5	-220	-220	-150
Drift6	-850	-750	-150
Drift7	-404	N.C.	N.C.
low. einzel lens	-300	N.C.	N.C.
mid. einzel lens	-300	N.C.	N.C.
upp. einzel lens	-300	N.C.	N.C.
Mesh	N.C.	-550	-550

Table 4.3: Highlighted are the voltage settings for the precision Penning trap extraction and ion optics at ISOLTRAP, used for ToF-ICR and PI-ICR in comparison for the new and the previous extraction schemes. The correction tube and the end cap are shown with the trapping potentials. For both ToF-ICR schemes, the ejection voltage applied to the end cap and the corrector electrodes is -19 V, whereas for PI-ICR these two elements are switched to -40 V.

Here N.C. represents elements that are not implemented in that specific construction. The comparison as shown in Table 4.3, represents the former ToF-ICR ejection scheme, as well as the previous electrostatic ion optics arrangement, between the new ToF-ICR voltage-set and the fast ejection scheme of PI-ICR for the ISOLTRAP setup. In contrast to the ToF-ICR technique, where a slow extraction procedure is necessary to obtain a large ToF effect, PI-ICR requires a fast extraction voltage, which allows a very fast ejection of ions from the area with a strong magnetic field gradient to minimize systematic effects, as described in Section 4.4. By use of these conditions in PI-ICR the ToF distribution of the ions between the trap center and detector is kept as narrow as possible $t_{axial} = 500$ ns. In this common procedure it is given that the ions, which travel with the same energy along the same magnetic field lines, observe the same systematic uncertainties. A subsequent discussion on this topic can be found in Section 4.4.1. Before describing further differences between the new ToF-ICR and PI-ICR settings, a brief summary of the issues of the old design, together with further improvements, is given in the following.

As an overview, the actual construction of the electrostatic optics, as well as the magnetic field strength at each position downstream to the detector is shown in Fig. 4.7:

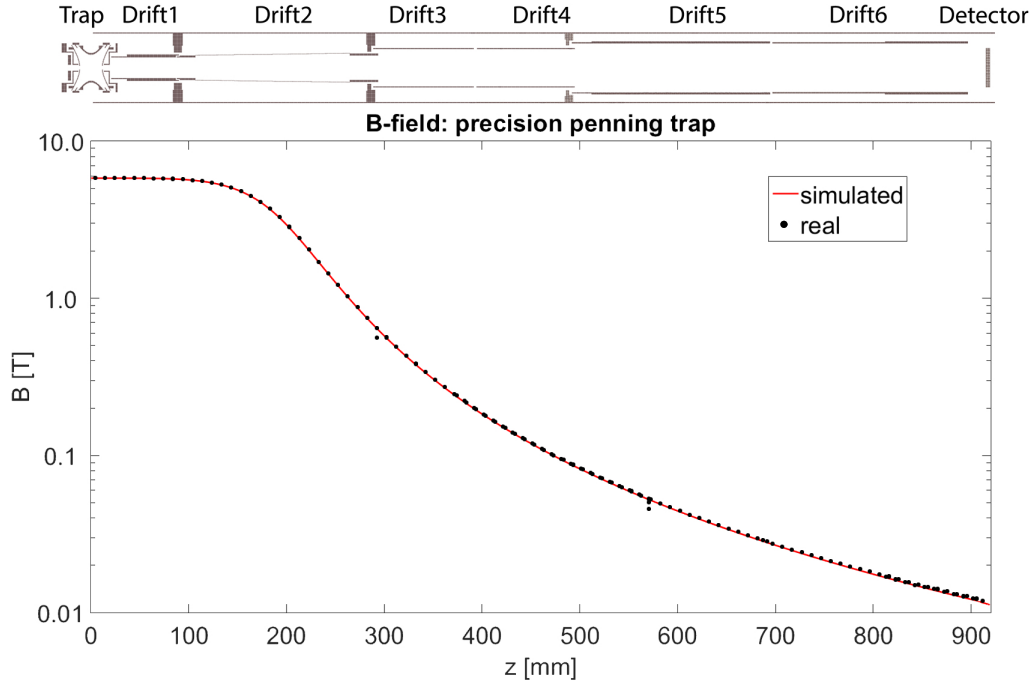


Figure 4.7: In the upper picture the precision Penning trap tower in its actual status at ISOLTRAP is illustrated. The tower contains the upper measurement Penning trap, the electrostatic tower electrodes, as well as the position-sensitive detector. After the ions are captured in the trap, the ions get excited with the ToF-ICR or PI-ICR cyclotron frequency scheme and projected with an ejection pulse onto the detector. Below those electrodes the magnetic-field strength B as a function of the distance z between the trap center and the detector is shown. The detector is located 913 mm downstream the Penning trap, surrounded by a residual magnetic field strength of only $\Delta B = 16$ mT. The dots represent real measured values [Ata16], where the line represents the simulated ones, by use of SIMION's Biot-Savart solver.

The processes in the handling of the ions by frequency excitations for PI-ICR, were described in Section 4.1. To preserve the right timings for each measurement cycle, to reduce systematic uncertainties, a well controlled and reliable trigger system has to be used. For this reason, a new set of frequency generators has been installed at ISOLTRAP, which are lined up in a certain formation. To reduce the uncertainties of the crucial excitation free accumulation time t_{accu} , two independent sets of frequency generators are used for each pattern obtaining almost the same settings. In case of the direct measurement scheme, explained in Section 4.1.2, the difference for both generator sets is the accumulation time t_{accu} . The accumulation time either occurs immediately after the π -pulse in case of pattern 1 or between the reduced

cyclotron excitation ν_+ and the converting π -pulse in pattern 2. The arrangement of the function generators at ISOLTRAP are described in the following schematic Fig. 4.8:

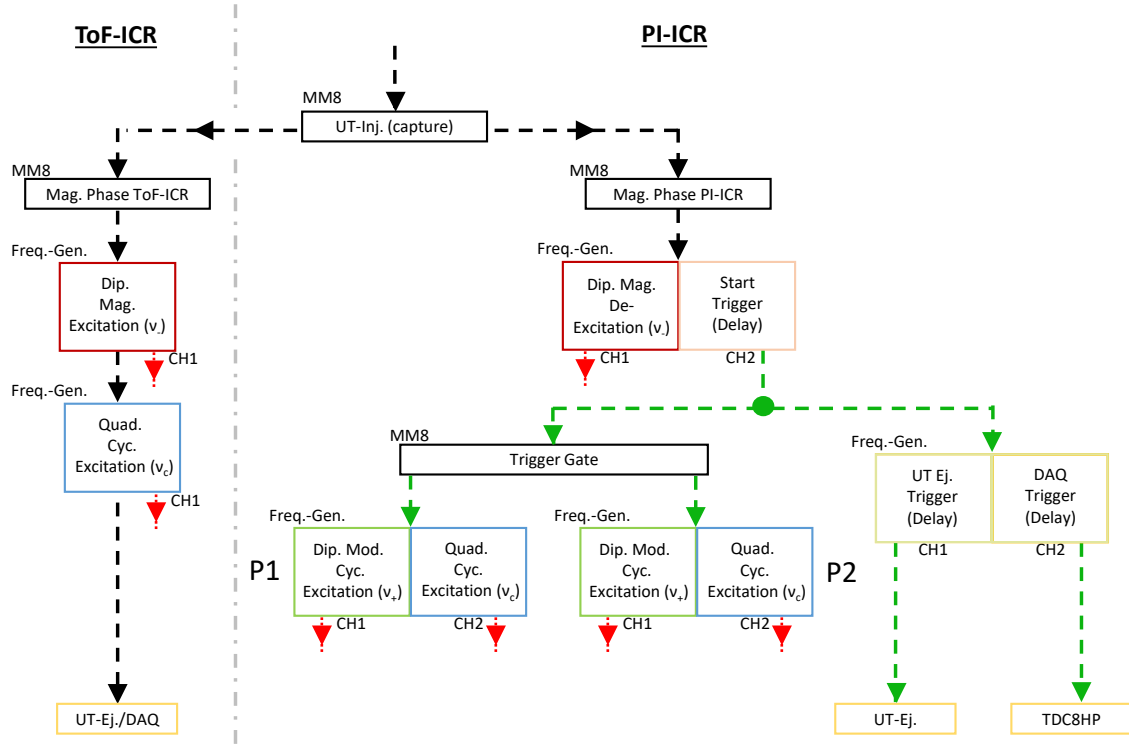


Figure 4.8: Illustrated are the new precision Penning trap (UT) cycle schemes for the ToF-ICR technique (left), as well as the PI-ICR technique (right), which are currently implemented at ISOLTRAP. Here, the parts which are controlled directly by ISOLTRAP's control system (MM8) are highlighted in black, where the elements controlled by a trigger pulse, generated by a frequency generator, are marked in green. The dipole and quadrupole frequencies which are applied to each ring electrode at the precision Penning trap segments by the frequency generator's output channels are illustrated with red labeled dots.

The schematic overview at the left hand side in Fig. 4.8 describes the actual path of the ToF-ICR measurement cycle for the precision Penning trap at ISOLTRAP. On the other hand the PI-ICR measurement cycle is described on the opposite side. Due to the different excitation and the different timing schemes among each other, the paths are highlighted in different colors and described below. The most important crucial point to consider between the two techniques is the initial magnetron motion and the precise control of the accumulation time t_{accu} for PI-ICR. Therefore, different frequency generators must be used to minimize these hampering effects. In both cases the ions get trapped after they were cooled in the preparation Pen-

ning trap. This step is managed by ISOLTRAP's trigger FPGA¹² pattern card (NI PCI-7811), which has a maximum timing resolution per channel, respectively, processing step, of $t_{res} = 25$ ns. In case of the ToF-ICR cycle, each step is managed by this FPGA card, as can be seen from the black dashed lines (Fig. 4.8). The ion's magnetron motion's phase in ToF-ICR is probed in a way that the ion trajectory is locked with the subsequent dipole excitation at the magnetron frequency ν_- , $\Delta\Phi = \pi/2$, to enhance the magnetron radius right from the beginning, as described in Section 2.2.4. Prior the ejection of the ions, the magnetron motion is converted into the reduced cyclotron motion ν_+ , as mentioned in Section 2.2.4. The different applied radio frequencies to the Penning trap ring segments are highlighted in both cycles by the red dotted arrow lines. In case of the ISOLTRAP's PI-ICR cycle on the right hand side of Fig. 4.8, the ion's magnetron motion is tuned in a way that the applied dipole de-excitation reduces the initial magnetron motion to its minimum with $\Delta\Phi = 3\pi/2$, as explained in Section 2.2.4. The FPGA pattern card in the PI-ICR mode, manages the uncritical slow magnetron phase timings, as well as the broad trigger gate. The broad trigger gate regulates the main trigger, which starts either the measurement procedure in pattern 1 (left P1 frequency generator) or pattern 2 (right P2 frequency generator), as stated in Section 4.1.1 I) and Section 4.1.2 II). The trigger paths managed by the main trigger frequency generator (Start Trigger), which are independent of the slower FPGA pattern card, are labeled with the green dashed arrow lines. The major step by moving the control of the dipole excitation and the quadrupole conversion, from the external FPGA pattern card to the internal frequency generator solution, as demonstrated with the lower part of the PI-ICR hardware setup in Fig. 4.8, reduces the crucial systematic accumulation time uncertainty from ~ 5 ns to < 200 ps by a factor 25. The FPGA card jitter will be explained by means of an example in Section 5.1. Further systematic studies with focus on the phase accumulation time and its precision are described in Section 4.4.8. Additional changes in the control system have adapted more automatic procedures to ISOLTRAP's control patterns. These changes provide an automatic cross-check ability between ions from ISOLDE or ISOLTRAP's alkaline ions, for MR-ToF MS, as well as for Penning-trap measurements, heading to a fully autonomous tuning procedure for ISOLTRAP in the near future.

As noted previously in Table 4.3, some additional electrostatic optics have been removed in the course of which the position-sensitive detector was lowered of about 23 mm [Kar17]. This led to a more homogeneous ion ejection path and limits the distortions of the ion spots observed in the previous construction, as shown in the following Fig. 4.9:

¹²FPGA is the acronym for Field Programmable Gate Array.

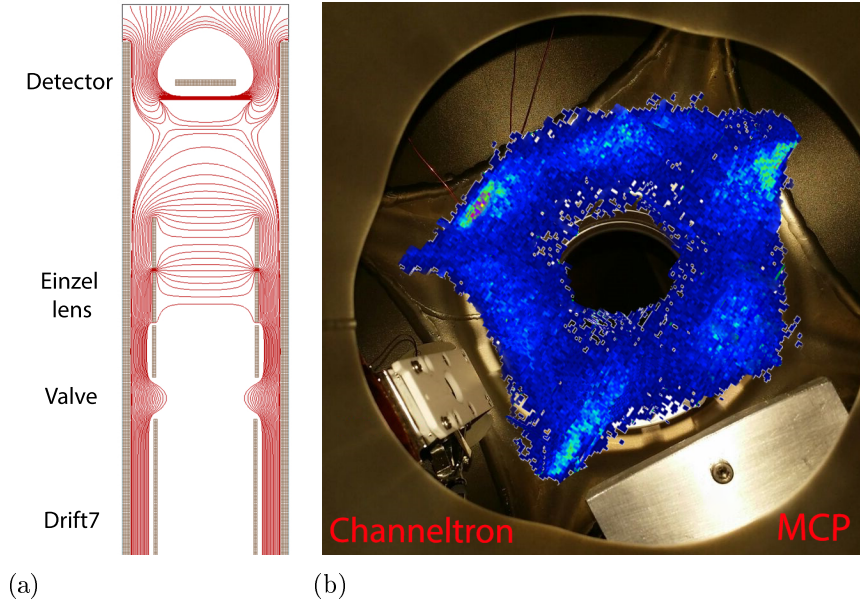


Figure 4.9: Both pictures represent the situation with the old ejection drift tube construction, including the einzel lens and the drift 7 element. Picture (a) illustrates the strong deformed electric field between the einzel lens and the detector. Thus, the ground potential of the six-way cross strongly influenced the trajectory of the ions, as can be seen in picture (b). The previous detection techniques the Channeltron (lower left corner), as well as the former MCP detector (lower right corner), strongly distorted the trajectory of the ions additionally. The real events, highlighted in blue in picture (b), represent the issue vividly.

The old drift elements, drift 7, as well as the einzel lense, were removed due to the fact that they caused distortions in the electrostatic field, as shown in Fig. 4.9 (a). For better visualization, an image of the six-way cross with the former Channeltron and the previously used MCP detector was superimposed by the ion events, marked in blue, which were recorded by the position-sensitive detector, as presented in Fig. 4.9(b). The picture was taken from above the Penning trap center. The main influences on the trajectory of the ions are generated by the strongly distorted electrostatic field over a distance of approx. 10 cm within the six-way cross, caused by a missing drift tube and the old detection systems. In order to avoid further distortion caused by the deep attractive potential of the position-sensitive MCP detector itself $MCP_{Anode} = -2.15$ kV, an additional honey-comb mesh with an ion transparency of $>75\%$ ¹³ was installed in front of the detector. Thus, the potential of the mesh can be adjusted to the potential of the last drift element, avoiding a strong electric field gradient, to mitigate the influences on the ion trajectory. Taking into account all changes made so far, the following picture shows the current status of PI-ICR at ISOLTRAP Fig. 4.10:

¹³The RoentDek MCP mesh, for the DLD40 delay-line detector is installed 8 mm below ISOLTRAP's MCP.

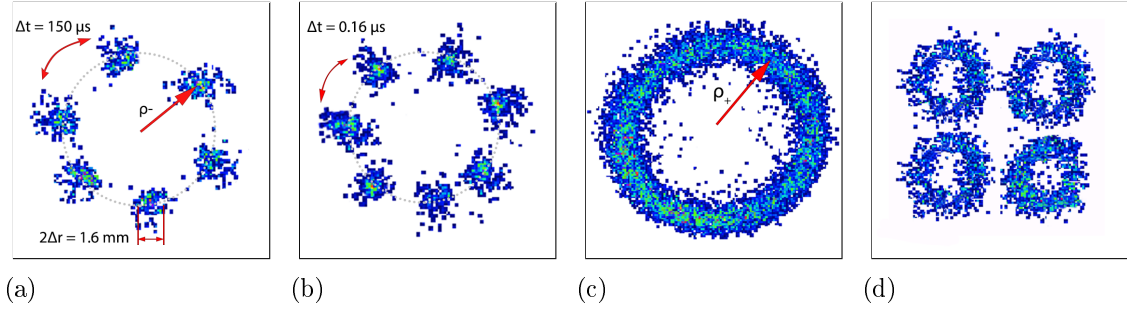


Figure 4.10: The measured projections of the different motions in PI-ICR for the isotope $^{85}\text{Rb}^+$ are shown. In (a) the full pure magnetron motion with ions ejected after an accumulation time in steps of $\Delta t_{\text{accu}} = 150 \mu\text{s}$, as well as in (b) the cyclotron motion in steps of $\Delta t_{\text{accu}} = 0.16 \mu\text{s}$ ejection spots after its full conversion, are presented. In (c) a direct projection of the pure cyclotron motion is visible, where in (d) an admixture of magnetron motion and cyclotron motion is illustrated at four different magnetron phases. See text for details.

The projection of the different ion motions towards the detector in Fig. 4.10 are recorded with $^{85}\text{Rb}^+$ by use of the settings of Table 4.3. On the left hand side in Fig. 4.10 (a), the magnetron motion is directly projected after the excitation with pattern 1, as mentioned in Fig. 4.3 (left). Here, the spatial distribution of the ions orbiting on a projected magnetron radius of $\rho_- = 10.5 \text{ mm}$, is $2\Delta r = 1.6 \text{ mm}$. Fig. 4.10 (b) represents the cyclotron motion projection after a full conversion to magnetron motion, with the excitation procedure of pattern 2 Fig. 4.3 (right). Without a conversion π -pulse as in Fig. 4.10 (b), the fast ion motion with an initial axial distribution of $t_{\text{axial}} = 1 \mu\text{s}$, is projected as a ring with the modified cyclotron motion radius of $\rho_+ = 10.5 \text{ mm}^{14}$, as shown in (c). Picture (d) represents the situation as an example if a non-zero magnetron motion at the beginning of the PI-ICR measurement cycle is remaining resulting into a mixture of both motions and a loss in precision and with it the wrong determination of the true frequency. The shown ion motions in Fig. 4.10, represented the current situation of PI-ICR at ISOLTRAP. The technique was implemented within this thesis and first systematic studies at ISOLTRAP to improve the reliability and the precision, is specified in the following Section 4.4.

¹⁴The ring structure on the modified cyclotron motion is generated by the initial axial motion, while the ions are spiraling with different time of flights around the magnetic field lines resulting in the projection as a ring structure, rather than a spot.

4.4 PI-ICR tuning strategy for ISOLTRAP and systematic uncertainties

Compared to the well understood standard ToF-ICR tuning procedure [Muk08], the focus of the tuning strategy for PI-ICR has changed. These changes are working comprehensively to each other, where the ToF-ICR technique will profit from the new studies and tuning strategies of PI-ICR. This section explains in more detail, the systematic uncertainties and their influencing impact on the daily measurement basis at ISOLTRAP. Most used formulas within this chapter are taken from [Eli14].

4.4.1 Minimizing the initial axial motion

The first step towards fine-tuning the precision Penning trap is the capture pulse of the pre-cooled ions. To obtain the minimal axial motion, the capture time has to be set to the correct timing. For this purpose a scan with the PI-ICR voltages and the ToF-ICR excitation procedure as explained in Fig. 4.8 can be used. The capture pulse will be scanned in the region, in which the ions are trapped in the precision Penning trap. In case of $^{85}\text{Rb}^+$, the range is determined, by $t_{\text{capture}} = 40 \pm 2 \mu\text{s}$, as presented in Fig. 4.11.

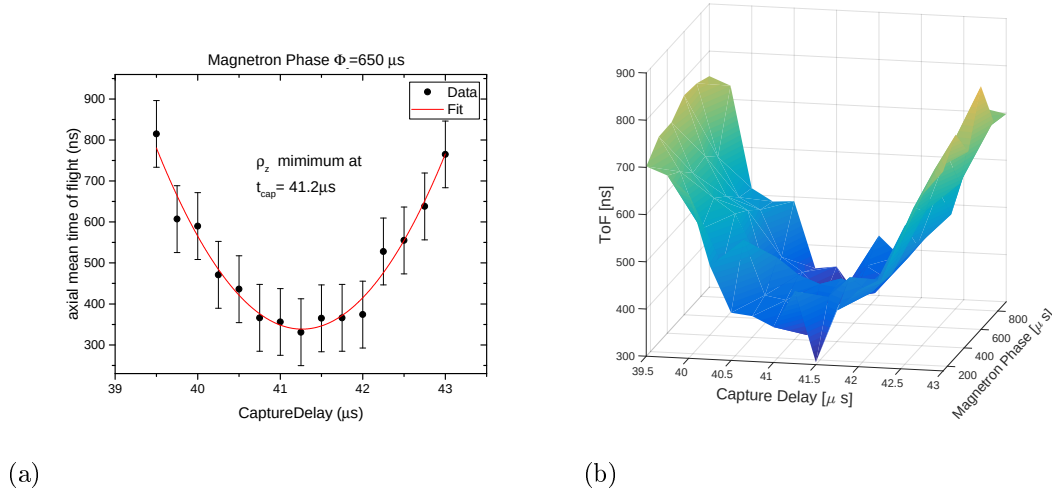


Figure 4.11: The picture represents the mean ToF distribution of the ions in conjunction with different axial energies, correlated as a function of different capture times of ISOLTRAP's precision Penning trap. In figure (a) the highlighted standard deviation of the mean axial ToF ion distributions for different capture times are marked for one specific magnetron phase $\Phi = 650 \pm 10 \mu\text{s}$, which is scanned for each capture time over the length of several axial motion periods. This procedure is repeated for several magnetron phases, completing the full round of the ions' axial motion around the clock, as shown in (b). The valley represents the mean best capture time of $t_{\text{cap}} = 41.2 \mu\text{s}$ at each magnetron motion phase step. Thus, the axial energy minimum, at a certain capture time, is equal within the uncertainties in the entire trap.

As shown in Fig. 4.11, the capture time will be scanned within a range of $40 \pm 2 \mu\text{s}$ in steps of $0.5 \mu\text{s}$, as in case of ^{85}Rb , representing the x-axis, where the y-axis highlights the mean ion ToF distribution, respectively axial energy. Thus, the minimum of the axial energy in each scan can be illustrated as the function of the capture time, as shown in Fig. 4.11 (a). In order to achieve this scanning method, each ion capture is followed by a magnetron phase scan, which is varied over several axial motion periods. This scan is followed by an additional magnetron dipole excitation and a cyclotron motion conversion as in the standard ToF-ICR method to determine the standard deviation of the ion ToF distribution. Furthermore should be mentioned that this procedure can be used to check whether the trap is well aligned with respect to the magnetic field lines. For this test, several points around the magnetron phase can be examined in the same way, as can be seen in the three-dimensional plot in Fig. 4.11 (b). The result of this scan shows that ISOLTRAP's precision Penning trap has a homogeneous standard deviation ToF distribution at each capture time within the uncertainties. Therefore, it is sufficiently accurate to examine the axial motion at any region on the preferred magnetron radius within the trap to set the correct trapping time, respectively, to reduce the axial ion energy to a minimum.

To be mentioned should be that the ToF-ICR excitation procedure used here for determining the minimum axial motion's phase could be replaced by the PI-ICR technique, as simulations have shown, Appendix A.5. Further tests under real conditions will soon provide further information.

4.4.2 Minimizing the initial magnetron motion and conversion admixture

After the correct capture time for the precision Penning trap was found to minimize the initial axial motion (Section 4.4.1), the influences of the initial magnetron motion and the improper conversion of the π -pulse have to be minimized.

Minimizing the initial magnetron motion

If a proper injection straight along the magnetic field axis into the precision Penning trap from the preparation Penning trap over a distance of 2.5 m is succeeded, the initial magnetron motion would be zero. One of the major issues at ISOLTRAP, which could lead to an initial magnetron motion, is therefore an angular displacement between the preparation Penning trap's magnetic field axis, with respect to the precision Penning trap's magnetic field axis. Thus, different masses will have different emerging trajectories from the preparation Penning trap, which means that the settings tuned for any mass will lead to an injection angle and an initial magnetron radius in the precision Penning trap. An additional possibility might be due to an improper cooling of the remaining motion in the preparation Penning trap, so that the ions are transported into the precision Penning trap off the central axis. At ISOLTRAP to compensate for these problems is to use a correct tuning method

and the use of ISOLTRAP's steerer elements in order to counteract for such offsets. Where in the case of the ToF-ICR technique the initial magnetron motion could have been used to start with an initial higher radial energy, however, in PI-ICR it is of importance to have no initial magnetron excitation, right before the application of the pi-pulse, to prevent an admixture of the reduced cyclotron motion and the initial magnetron motion. Due to this fact, the magnetron de-excitation as explained in Section 4.1.2, was introduced to de-excite the ions with a dipole pulse at the magnetron frequency with respect to the ions' motion phase of $\Delta\Phi_- = 3\pi/2$, (see Section 2.2.4). To accomplish the smallest initial magnetron motion, the magnetron phase has to be scanned as shown in Fig. 4.12 (a):

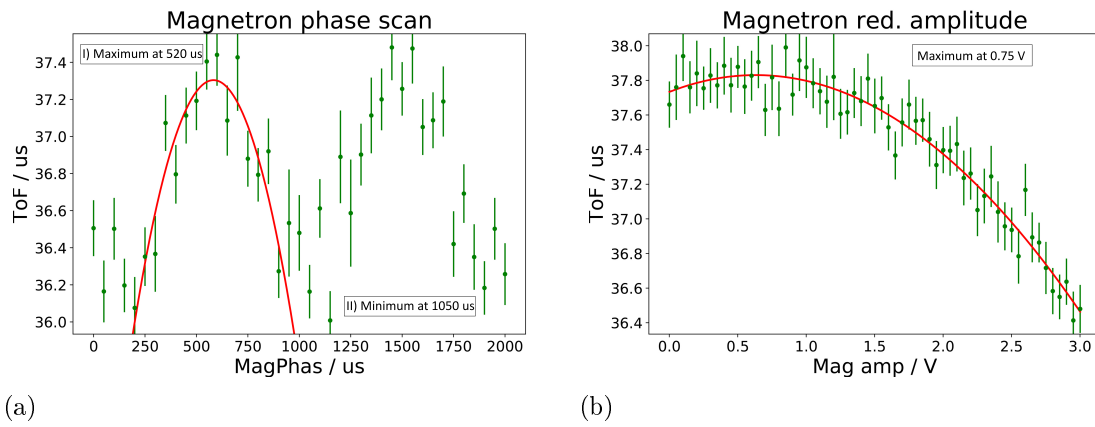


Figure 4.12: On the left hand side (a), the results of the ToF-ICR cyclotron conversion procedure is shown at different ion magnetron motion phases with their corresponding ion ToF. Where at position (I), the highest flight time marks the phase $\Delta\Phi_- = 3\pi/2$, which determines the correct magnetron phase for the PI-ICR mode, respectively, in position (II) the correct magnetron phase for the ToF-ICR mode. On the right hand side (b) a scan of the magnetron amplitude is shown in order to obtain the smallest initial magnetron motion with the correct de-excitation pulse after the correct magnetron phase has been set.

The shown Fig. 4.12 can be obtained with the ToF-ICR excitation scheme, where first the magnetron phase is scanned up to a certain moment, which is subsequently followed by a magnetron dipole excitation and the cyclotron conversion. If the flight time of the ion is longer, the ion had less energy in the radial plane. The dipole excitation reduces the energy in the radial plane, the more the ion approximates the phase difference between the ion on the magnetron motion and the excitation field of $\Delta\Phi_- = 3\pi/2$, as can be seen in Fig. 4.12 (a)(I) for a magnetron phase delay 520 μs . This phase contributes the least to the energy increase in the radial plane. Whereas the more the ion reaches the magnetron phase difference, with respect to the ion of $\Delta\Phi_- = \pi/2$, the more energy is available. Thus, resulting in a shorter flight time as highlighted at position Fig. 4.12 (a)(II). Therefore, the position for a small magnetron motion, which is of importance for PI-ICR is marked at position

Fig. 4.12 (a)(I). In case of ToF-ICR, position Fig. 4.12 (a)(II) with $1050 \mu\text{s}$ ¹⁵, indicates the correct phase for the highest initial magnetron motion.

After the magnetron phase for PI-ICR is determined, the magnetron amplitude for a certain number of excitation cycles (three cycles at ISOLTRAP) has to be scanned to achieve the smallest initial magnetron motion. Thus, the magnetron amplitude is scanned, as demonstrated in Fig. 4.12 (b), at the correct magnetron phase for PI-ICR, by use of the ToF-ICR excitation pattern. The longer the ToF value, the smaller the initial magnetron motion due to the reduced energy in the radial plane. Using the magnetron phase delay of $520 \mu\text{s}$ leads to a magnetron amplitude of $U_{\rho-} = 0.75 \text{ V}$.

An alternative to the ToF determination procedures for both scanning methods, as shown in Fig. 4.12, would be to compare the scans with the ion spot size projected onto the position-sensitive detector. In both scans, the smallest initial magnetron motion is determined by the smallest spatial ion spot distribution.

The right quadrupole conversion amplitude

In the third step, after minimizing the initial axial energy and the initial magnetron motion, the correct quadrupole conversion π -pulse amplitude has to be determined. Prior each voltage scan step the dipole excitation at the reduced cyclotron frequency ν_+ excites the ion to a certain cyclotron radius of about $\rho_+ = 0.6 \text{ mm}$. Subsequently, the quadrupole conversion is then applied at the correct free cyclotron frequency ν_c , while changing the amplitude of the cyclotron excitation in each scan step. The amplitude scan, with a fixed excitation time, here, $T_{ex} = 1.4 \text{ ms}$ is highlighted in the following Fig. 4.13:

The presented π -pulse amplitude scan in Fig. 4.13 (a), is highlighted with three intermediate amplitude steps (I, II, III). As in case of the procedure in the previous subsection, the time of flight information of the different excitation phases, respectively, quadrupole conversion amplitudes U_{quad} , can be compared to each other. Here, the longest ToF corresponds to one full conversion, from the pure reduced cyclotron motion into the pure magnetron motion, as it should be the case for a correct PI-ICR measurement. Another possibility to determine the correct π -pulse amplitude is to reach the smallest ion spatial spot distribution on the position-sensitive detector, as demonstrated with real ion events on the right hand side Fig. 4.13 (b) (I, II, III). In case of Fig. 4.13 (b)(I) the conversion amplitude is $U_{quad} = 0 \text{ V}$, so that the pure reduced cyclotron motion is projected towards the detector, which leads to a projection of a ring. At position Fig. 4.13 (b)(II), the amplitude for a given excitation conversion time was not sufficient enough to convert the pure reduced cyclotron motion into the pure magnetron motion, an admixture of both motions remains. A complete motion conversion, as can be seen in Fig. 4.13 (b)(III), is achieved here at $U_{quad} = 2.15 \text{ V}$. It should be mentioned that the pure reduced cyclotron motion seems to be a ring after its projection, where in the trap it is located at the same position as if the ions are on the magnetron motion. The full ring at ISOLTRAP

¹⁵If no initial magnetron motion exists, the magnetron phase scan would show no ToF variation.

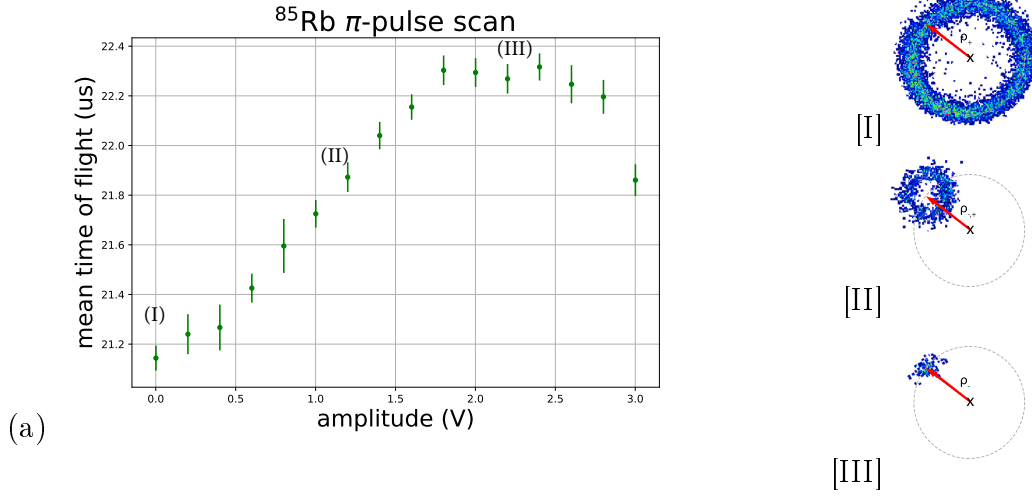


Figure 4.13: Figure (a) shows the quadrupole conversion amplitude scan with its corresponding mean time of flight behavior of the ions from a pure cyclotron motion (I), over an admixed state of magnetron and reduced cyclotron motion (II) to a pure magnetron motion (III). On the right hand side, the position-sensitive detector projections for the different (I, II, III) conversion amplitudes are visualized. The conversion time for each excitation is kept constant at $T_{conv} = 1.4$ ms, as in case for a singly charged $^{85}\text{Rb}^+$ ion.

can only be visualized if an initial axial motion larger than $t_{axial} > 1 \mu\text{s}$ is remaining, otherwise the ring is less pronounced. In order to demonstrate this effect, the axial energy has been increased. Since the current ISOLTRAP design does not allow the axial motion to be reduced to an acceptable minimum where direct projection of the cyclotron motions as a spot on the position-sensitive detector is possible, it is difficult to determine the reduced cyclotron frequency directly and is therefore not currently used for PI-ICR measurements.

The procedures discussed within this subsection have to be determined for each mass that have to be measured. Probing the magnetron phase to be able to minimize the initial magnetron motion, with the correct de-excitation amplitude, is the most crucial point, due to the previously mentioned transport issues between the preparation Penning trap and the precision Penning trap. The scan of the correct capture time, as well as the evaluation of the correct conversion amplitude for the quadrupole conversion pulse are less important as they can be scaled very accurately from one mass to another.

4.4.3 Harmonizing the electric and magnetic trapping field

The electric field, as well as the magnetic field imperfections, were introduced in Section 2.2.3. However, how the effects of the individual perturbation can be determined using the standard ToF-ICR and the novel PI-ICR method is discussed in

this section.

Magnetostatic imperfections

The magnetic field perturbations determined in Eq. (2.34), cause a shift of the cyclotron frequency as a function of even powers of the ion distance with respect to the trap center. This effect will be used as an advantage to probe different trapping regions with the cyclotron frequency. Thus, the magnetic field homogeneity over a specific trap volume can be determined. In case of the standard ToF-ICR technique the ions are captured over a certain capture time scan range. For each capture time a full ToF-ICR resonance of $T_{ex} = 1$ s is performed. This procedure provides the free cyclotron frequency shift $\Delta\nu_c^{mag}$, whereby the result of the capture time with the least axial motion is used as the reference free cyclotron frequency $\nu_{c,ref}$. The smaller the free cyclotron frequency shift with respect to the center frequency becomes, the more homogenous the magnetic field region is. To compensate the inhomogeneities in the magnetic field, ISOLTRAP's precision Penning trap is equipped with a room temperature current regulated z^2 shim coil. By use of the correct compensation current, here, $I_{shim} = 283$ mA, the uncertainty to the magnetic field becomes less than $\Delta B/B_0 < 1.2 \cdot 10^{-8}$, within a capture time range of $UT_{cap} = 41.2 \pm 2 \mu\text{s}$ around the center, as can be seen in Fig. 4.14. A more detailed description can be found in [Muk08].

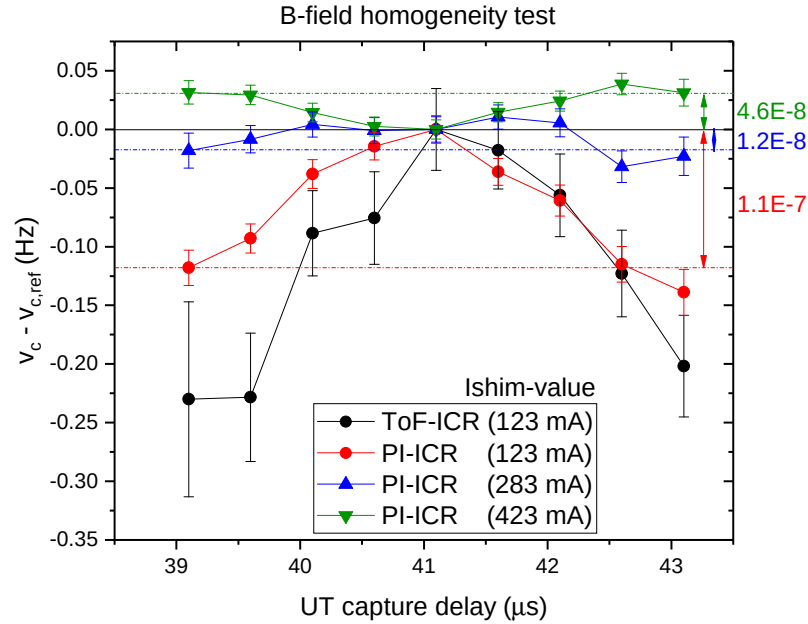


Figure 4.14: Demonstrated are the effects of different z^2 -shim compensation coil current values (I) 100 mA (II) 283 mA (III) 350 mA and their effect on the magnetic field homogeneity within a certain trap volume. Only one ToF-ICR measurement is compared to the PI-ICR measurements for the sake of clarity. See text for further details.

In the following, a novel procedure for the investigation of magnetic field homogeneity at ISOLTRAP with the aid of the PI-ICR method in the direct measurement procedure (Fig. 4.3) for the determination of the free cyclotron frequency ν_c shifts is presented. The capture time around the center with the smallest initial axial energy is scanned, as previously described with the ToF-ICR technique (Section 4.4.1). For each capture time, the excitation (pattern 1), as well as (pattern 2) are applied to determine the free cyclotron frequency. This procedure is more than six times faster as in case of the ToF-ICR technique. The result of the explained procedure can be seen in Fig. 4.14, where the standard ToF-ICR (black curve) and the novel PI-ICR procedure are compared to each other, with the effect of different shim coil correction currents.

With *SIMION*, the ion axial deflection within the precision Penning trap at different capture times can be simulated, as demonstrated in Appendix A.4. Thus, resulting in an axial deflection of $z = 1$ mm at $41 \mu\text{s}$ and $z = 9$ mm at $39 \mu\text{s}$, respectively, $43 \mu\text{s}$, for $^{85}\text{Rb}^+$.

Now that the overall magnetic field homogeneity tuning procedure is clarified, a more detailed view will be given for the frequency shifts induced by magnetic perturbations. To give a brief overview, the previously used equations in Eq. (2.34) describing the magnetron frequency shift $\Delta\nu_-$ and the reduced cyclotron frequency shift $\Delta\nu_+$ are rewritten in a slightly different notation to guide the reader through the section, in particular for the determination of the systematic uncertainties at ISOLTRAP's precision Penning trap, using the PI-ICR technique. By use of the equations given by [Ket14] the second order of non-zero perturbations act as a frequency shift to:

$$\Delta\nu_+ \approx \frac{B_2}{B_0} \cdot \frac{\nu_+}{2} \cdot \left(z^2 - \rho_+^2 - \rho_-^2 \left(1 + \frac{\nu_-}{\nu_+} \right) \right), \quad (4.18)$$

for the cyclotron frequency, where the deviation for the magnetron frequency is represented by:

$$\Delta\nu_- \approx -\frac{B_2}{B_0} \cdot \frac{\nu_-}{2} \cdot \left(z^2 - \rho_+^2 \left(\frac{\nu_+}{\nu_-} + 1 \right) - \rho_-^2 \right). \quad (4.19)$$

Here, B_0 and B_2 represent the first two non-zero series expansion coefficients of the magnetic field strength $\mathbf{B}$, as introduced in Eq. (2.33). The parameter ρ defines the ion's location in the radial-plane with respect to the trap center, where z determines the axial amplitude of the ion's motion. In case of ISOLTRAP, the precision Penning trap has its center magnetic field strength of $B_0 = 5.9$ T. With the help of the determined uncertainty of the magnetic field $\Delta B/B_0 < 1.2 \cdot 10^{-8}$, in Fig. 4.14 and a simulated maximal axial ion deflection of $z = 9$ mm, as shown in Appendix A.4, the B_2 component of the precision Penning trap at ISOLTRAP can be calculated to be $B_2 = 1.7 \cdot 10^{-9}$ T/mm². By use of Eqs. (4.18) and (4.19) the following assumption can be made:

$$\Delta\nu_+[Hz^{-1}] = 2.8 \cdot 10^{-10} \cdot \frac{\nu_+}{2} \cdot \left(z^2 - \rho_+^2 - \rho_-^2 \left(1 + \frac{\nu_-}{\nu_+} \right) \right) [mm^{-2}], \quad (4.20)$$

$$\Delta\nu_-[Hz^{-1}] = -2.8 \cdot 10^{-10} \cdot \frac{\nu_-}{2} \cdot \left(z^2 - \rho_+^2 \left(\frac{\nu_+}{\nu_-} + 1 \right) - \rho_-^2 \right) [mm^{-2}]. \quad (4.21)$$

After clarifying the possible frequency shifts caused by magnetostatic perturbations of the precision Penning trap magnetic field, a representative example will be used to show the influence of this interference on the radial frequencies within the uncertainties. Considering the minimum axial amplitude $z = 1$ mm, as demonstrated in the simulation Appendix A.4 and the achievable maximum of the magnetron motion $\rho_- = 1.2$ mm for a singly charged $^{85}\text{Rb}^+$ ion, which performs an almost pure magnetron motion in the trap with a minor remaining admixture of the reduced cyclotron motion of $\rho_+ = 0.05$ mm, the resulting values are as follows:

$$\Delta\nu_+[Hz^{-1}] = -0.5 \cdot 10^{-4} \cdot \rho_-^2 [mm^{-2}], \quad (4.22)$$

$$\Delta\nu_-[Hz^{-1}] = 3 \cdot 10^{-7} \cdot \rho_-^2 [mm^{-2}]. \quad (4.23)$$

Using the maximum achievable magnetron radius at ISOLTRAP's precision Penning trap of $\rho_- = 1.2$ mm, the magnetron shift can be considered zero by means of Eq. (4.23), as well as the cyclotron-frequency shift Eq. (4.22), which does not exceed 0.1 mHz. The later frequency shift is not detectable within the current uncertainties on the detector for phase-accumulation times smaller than $t_{\text{accu}} < 10$ s. Thus, both frequency shifts, respectively, the angular position shift, caused by magnetic field inhomogeneities are relatively small and can be neglected within the current uncertainties.

Electrostatic imperfections

The magnetic field imperfection investigations suggest that the main source of systematic uncertainties affecting the radial ion motions can be considered to be the deviation of the electric trapping potential from the ideal quadratic form. The two leading non-quadratic series expansion perturbations which contribute the most, are the octupole C_4 and dodecapole C_6 terms, as previously introduced in Section 2.2.3. The non-quadratic terms caused by the trapping potential perturbations, lead to smeared image spots on the position-sensitive detector, which increases the uncertainty of the spatial distribution by applying larger motion radii, as shown later in Fig. 4.15 and demonstrated in [Eli14]. Therefore, the resolution is limited so that the accuracy is strongly correlated with these effects. Due to the fact that the cyclotron motion is affected by residual buffer-gas interaction, which additionally leads to a smearing of the image spot, the magnetron motion is privileged for the tuning of the Penning trap's harmonicity for the PI-ICR method. The above mentioned trapping-potential coefficients result into a relative shift of the magnetron frequency [Ket14] to:

$$\frac{\Delta\nu_-}{\nu_-} \approx \left[\frac{3}{2} \cdot \frac{C_4}{d^2} + \frac{45}{4} \cdot \frac{C_6}{d^4} \cdot z^2 \right] \cdot \rho_-^2 + \left[\frac{15}{8} \cdot \frac{C_6}{d^4} \right] \cdot \rho_-^4 + C. \quad (4.24)$$

Eq. (4.24) can be used for reduced cyclotron radii which are much smaller than the magnetron radius ρ_- . The constant C contains the magnetron radius ρ_- independent terms. When utilizing the ISOLTRAP values of the trap dimension, the following relation can be obtained:

$$\begin{aligned} \frac{\Delta\nu_-}{\nu_-} &\approx [14.3 \cdot 10^{-3} \cdot C_4 + 1 \cdot 10^{-3} \cdot C_6 \cdot z^2] \cdot \rho_-^2 + 1.7 \cdot 10^{-4} \cdot C_6 \cdot \rho_-^4 + C \\ &= C_4^* \cdot \rho_-^2 + C_6^* \cdot \rho_-^4 + C. \end{aligned} \quad (4.25)$$

To probe the E-field homogeneity of the precision Penning trap, two different procedures are available at ISOLTRAP, either with the common ToF-ICR technique or the novel PI-ICR technique. To give an overview both applications will be shortly presented in the following.

Considering the case of the standard ToF-ICR technique, the ions are captured at different times, where for each capture time the ToF-ICR excitation is applied. The slight difference to the usual ToF-ICR excitation shown in Section 2.2.7 is that the magnetron dipole excitation is followed by an additional dipole excitation sampled around the reduced cyclotron frequency ν_+ . The excited ions orbiting on a pure magnetron motion get an admixture of the reduced cyclotron frequency, which leads to the effect that different volumes in the trap are probed. The homogeneity is thereby determined by the different obtained reduced cyclotron frequencies in comparison to the center reduced cyclotron frequency, which is considered to be the capture time with the smallest initial axial energy. This ToF-ICR based technique is a time consuming tuning procedure.

By contrast, using the harmonization scan of the trap volume with the novel PI-ICR method saves the time that would otherwise be required to achieve the time-of-flight effect when scanning different frequencies. In the case of the harmonization procedure with PI-ICR, it is sufficiently accurate to capture the ion at a fixed capture time, where the ions get the least axial energy. This prevents the smearing of the spots on the detector due to the axial motion, as mentioned in Section 4.4.1. The ion capture is followed by a dipole excitation at the magnetron frequency ν_- to induce energy into the radial-motion. The magnetron motion as a probe for the electric field imperfections is the privileged one, because it is less influenced by remaining residual buffer-gas interactions, as in case of the cyclotron frequencies, which will be shown in Section 4.4.4. To probe the whole volume from the center onwards to the maximum achievable radius of $\rho_- = 1.2$ mm, the ions getting excited stepwise over several magnetron amplitudes, e.g. in $U_{step} = 0.1$ V steps, respectively, $\rho_{-,step} = 0.03$ mm per amplitude step, as shown in Fig. 4.15:

The ions probe the volume over a certain accumulation time, here $t_{accu} = 600$ ms, before they get ejected towards the position-sensitive detector. This fixed accumulation time preserves the angle in a linear behavior with respect to the linear increasing magnetron radii steps, as long as the trapping volume is not perturbed by the electrical field imperfections. The ideal harmonized trap achieves a linear

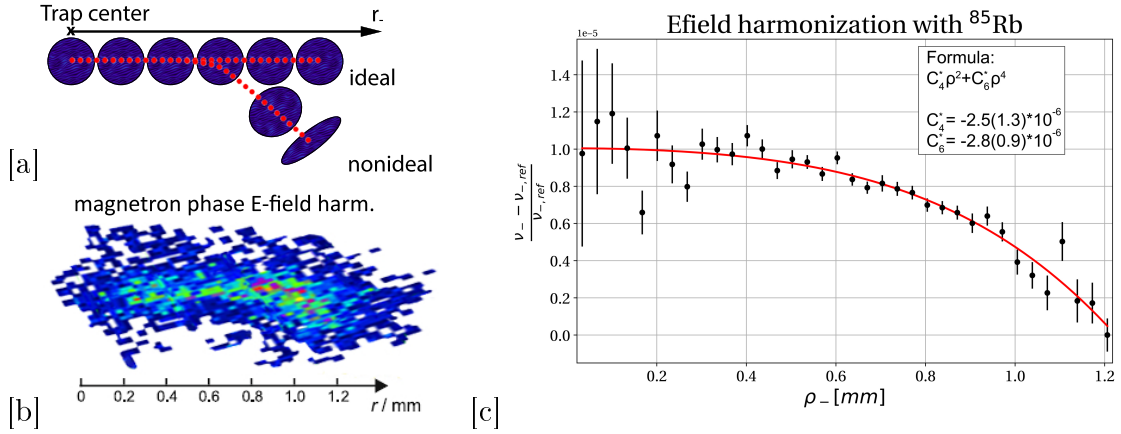


Figure 4.15: In the upper left picture (a) a sketch of the ideal and non-ideal harmonized Penning trap is shown. In case of an harmonized trap the ion trajectory increases linearly, proportional to the magnetron radius, where in case of the perturbed homogeneous electric field the trajectories are distorted. In picture (b) real ion events are recorded and represent the harmonization process of the precision Penning trap with the PI-ICR technique. The fit to the obtained values in (b) is represented in (c), determining the octupole C_4^* and dodecapole C_6^* perturbations. For further information see text and [Eli14].

trend at ISOLTRAP up to a radius of $\rho_- = 0.6$ mm, as presented in Fig. 4.15. The ideal case would be the harmonization over the whole accessible trap volume $\rho_- = 1.2$ mm. Further harmonization in smaller trapping potential tuning steps have to show if this is achievable. In the case of a non-harmonized trap volume, the frequency and ideal trajectory of the ions change and the isotropic spatially distributed ions begin to distort in the radial plane, as demonstrated with the non-ideal case in Fig. 4.15 (a). As an example, the harmonized precision penning trap with real ion events is shown in Fig. 4.15 (b). The degrees of freedom to harmonize a Penning trap at ISOLTRAP, are the end cap electrodes, the correction ring electrodes, as well as the correction tube voltages, as introduced in Section 2.2.3. In order to characterize which correction voltages are best suited for the harmonization, a full-scale magnetron amplitude scan must be performed. The recorded values from the scan can be fitted as shown Fig. 4.15 (c), using the magnetron radius $\rho_- = 1.2$ mm as the reference magnetron frequency to create the residual magnetron frequency of each magnetron amplitude. The fit yields the octupole $C_4^* = -2.5(1.3) \cdot 10^{-6} \text{ mm}^{-2}$ and dodecapole $C_6^* = -2.8(0.9) \cdot 10^{-6} \text{ mm}^{-4}$ perturbation terms, as introduced in Eq. (4.25).

4.4.4 Residual gas interaction

Since the pressure gauge with its strong magnet must be further away from the superconducting magnet of the Penning trap, as well as the limited space near the precision Penning trap, the exact pressure in the trap center is not directly accessible by a pressure gauge. This obtained pressure value is therefore only a guideline of the

pressure 1 m further away from the trap center. To determine the existing pressure in the trap center nevertheless, the ions as an efficient direct probe to the remaining rest gas can be used and determined by the PI-ICR technique. As the ions have a higher sensitivity to rest gas interaction in the reduced cycle motion, as shown in the preparation Penning trap procedures Section 2.2.5, the ions are converted with pattern 2, as shown in Fig. 4.3 (right), where the ions remain longer on the reduced cyclotron motion. In order to obtain the average number of ion collisions with residual buffer gas during the phase-accumulation time t_{accu} , the total distance of the ion's flight path in the Penning trap can be divided by their mean free path. As an example, a singly charged $^{85}\text{Rb}^+$ ion, stored over $t_{accu} = 1$ s, travels $d_{orbit} = 6.7$ km if it orbits on a $\rho_- = 1$ mm radius on the reduced cyclotron motion or $d_{orbit} = 7$ m with the velocity of the magnetron motion in vicinity of a magnetic field of $B_0 = 5.9$ T. Considering the mean free path of this mass in air at a pressure of 10^{-7} mbar is a few km, where in case of residual helium pressure of 10^{-8} mbar the mean free path exceeds ten km [Rot12]. In comparison with the travel distance on the magnetron motion the interaction with the residual gas is neglectable for the damping, respectively, the smearing of the spatial distributed spot. A second conclusion which can be drawn is that accumulation times below $t_{accu} < 1$ s can be considered to be without influences on the ion motions, if the pressure is well below 10^{-8} mbar.

To describe in detail the behavior of the ions, the following drawing Fig. 4.16 gives a rough estimate of the situation at the moment the ions interact with residual gas while they perform a pure reduced cyclotron motion: The ion which interacts with remaining buffer-gas molecules or atoms decreases its velocity from the initial v_+ to the velocity after the interaction v'_+ , with a certain scattering angle α . Thereby, the ion's initial reduced cyclotron radius ρ_+ is decreased by a certain amount to ρ'_+ , whereby the magnetron radius ρ'_- increases, due to its metastable nature. These processes are described more detail in Section 2.2.5, as well as in [Geo11]. This admixture of the magnetron motion results into the smearing of the ion spatial spot distribution and is therefore a limitation of the achievable maximum resolving power. The correlation between the reduced cyclotron and magnetron radius can be described as in the following:

$$\rho'_- = \sqrt{(\rho_+)^2 + (\rho'_+)^2 - 2\rho_+\rho'_+ \cos \alpha}. \quad (4.26)$$

For simplicity it can be assumed that the ion loses only the energy after an interaction with the residual buffer gas, without changing the angle, thus, without changing the direction in motion in the radial plane. By this Eq. (4.26) is reduced to:

$$\rho'_- = \rho_+ - \rho'_+ = \rho_+ \cdot \left(1 - \frac{\rho'_+}{\rho_+}\right) = \rho_+ \cdot (1 - k). \quad (4.27)$$

With Eq. (4.40) it is possible to estimate an average magnetron radius of the ion after a residual gas interaction by determining the damping factor k . By use of the viscous drag force, which counteracts the ion's motion as introduced in Eq. (2.48), the

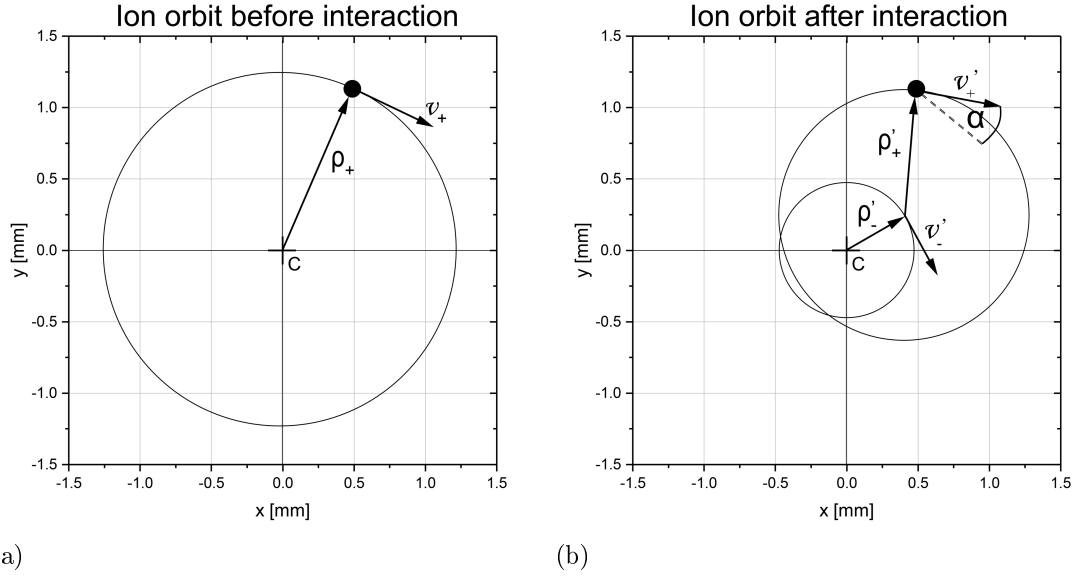


Figure 4.16: On the left hand side in picture (a), the ion performs an undisturbed pure reduced cyclotron motion on a reduced cyclotron motion radius of ρ_+ , with its drift velocity v_+ . On the other hand, in (b) the ion is represented with the situation after an interaction with residual gas. Thereby, the ion changes its velocity to v'_+ , with a corresponding scattering angle α , a reduction of the reduced cyclotron radius ρ'_+ , thus an increase of the magnetron radius ρ'_- . C represents the trap center. [Eli14].

damping constant of the cyclotron radius can be written as $\delta = \gamma_+$ [Kre08; Geo11]. The resulting relation to determine the damping factor $k = e^{-\gamma_+ t_{accu}}$ [EAM88] can then be used to estimate the real remaining pressure in the precision Penning trap as shown in the following Fig. 4.17:

Taking into account the damping constant $\gamma_+ = 2.8 (14) \cdot 10^{-2} \text{ s}^{-1}$, obtained by the fit in Fig. 4.17 (a) and the ion mobility, the pressure in the precision Penning trap can be determined, as described in Eq. (2.2). Where the ion mobility for a singly charged $^{85}\text{Rb}^+$ ion in water vapor is $K_{0,H_2O} = 0.7 \cdot 10^{-4} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$, respectively, in helium $K_{0,He} = 21.2 \cdot 10^{-4} \text{ m}^2\text{s}^{-1}\text{V}^{-1}$ [Eli84], as defined by Blanc's law [Bla08; Pet86]. Thus, if all motion damping had been on water vapor, the corresponding pressure would be $p_{H_2O} = 1.6 \cdot 10^{-9} \text{ mbar}$, where, if the damping had been on helium atoms, the pressure would result to $p_{He} = 5 \cdot 10^{-8} \text{ mbar}$, as expected at ISOLTRAP. From this it can be concluded that the main contribution to the ion interaction at ISOLTRAP is due to the remaining helium atoms escaping from the preparation Penning trap. By this assumptions ISOLTRAP's resolving power is limited to a few 10^7 for singly charged ions of mass 85 u. In order to achieve an even higher resolving power, as it would be possible with long-lived nuclides, the precision Penning trap tower with its fragile elements could be gently baked out or, as a second option, the helium pressure in the preparation Penning trap could be reduced. However, the later procedure can lead to a deterioration of the ion motion cooling, which could result to a counterproductive increase of the radial and axial motions.

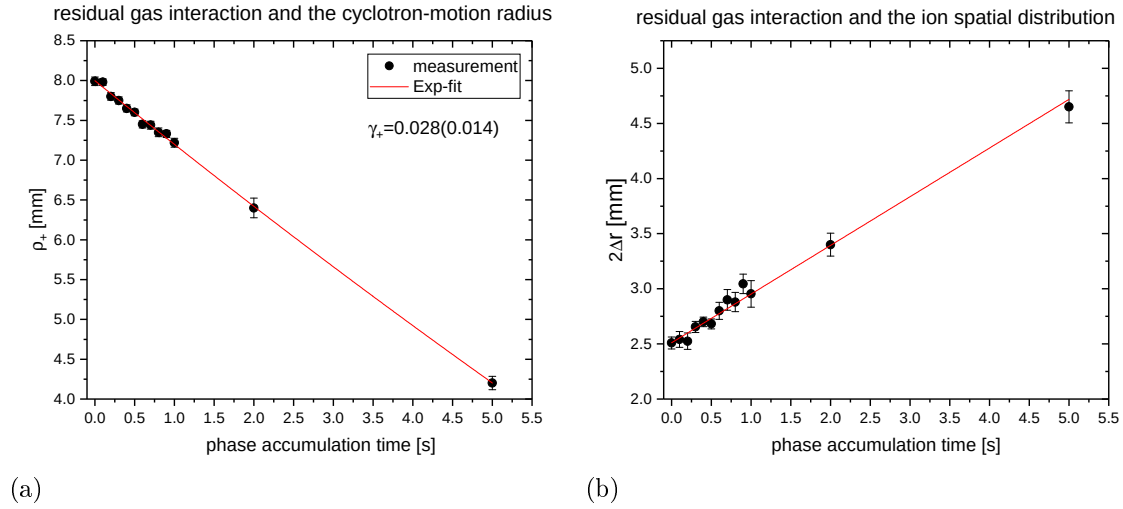


Figure 4.17: Both figures show the behavior of the trapped ions, which interact with the remaining buffer gas in the precision Penning trap. On the left hand side, the reduced cyclotron motion radius as a sensitive probe for ion interaction is compared to different accumulation times, representing a decreasing trend. Where the spot size increases at the same time due to the residual gas interaction, as shown on the right hand side, as an effect of the increasing proportion of the magnetron motion. The red line corresponds to the exponential fit to the data, as mentioned in [Eli14].

Other effects that influence the resolving power, respectively, the precision, are explained below.

4.4.5 Disruptive factors of the frequency to be measured

In the previous Section 4.4.4, the resolution limits were explained due to the residual gas interaction, while now discussions follow that keep the focus in situations that influence the excitation frequencies. This chapter will cover the points of influences introduced by non-simultaneous measurements of the reference and the final phase Section 4.4.5, voltage fluctuations Section 4.4.5, linear and angular displacement with respect to the trap's electrical and magnetic field symmetry axis Sections 4.4.6 and 4.4.6, as well as the implied uncertainties caused by a non-perfect interconversion between different motions Section 4.4.7.

Penning trap voltage fluctuations

One of the most significant effects that affect the ion spatial distribution of the phase spots is next to the rest-gas interaction, as shown in Section 4.4.4, the voltage instabilities of the trapping potential of the precision Penning trap. In the following, the influences at the magnetron and the reduced cyclotron frequency are discussed separately. The discussion within this subsection will be without the magnetic field instabilities, due to its minor influence at the spatial spot distribution. The average

fluctuations over a phase-accumulation time t_{accu} at the specific measurement time t are determined by [Eli14]:

$$\bar{\nu}_-(t) \approx \frac{1}{4\pi d^2} \cdot \frac{1}{B} \cdot \frac{\int_t^{t+t_{accu}} U(t') dt'}{t_{accu}} = \frac{1}{4\pi d^2} \cdot \frac{1}{B} \bar{U}(t, t_{accu}), \quad (4.28)$$

for the averaged magnetron motion $\bar{\nu}_-$ and

$$\bar{\nu}_+(t) \approx \nu_c - \frac{1}{4\pi d^2} \cdot \frac{1}{B} \cdot \frac{\int_t^{t+t_{accu}} U(t') dt'}{t_{accu}} = \nu_c - \frac{1}{4\pi d^2} \cdot \frac{1}{B} \bar{U}(t, t_{accu}), \quad (4.29)$$

for the average reduced cyclotron motion $\bar{\nu}_+(t)$, obtained by the subtraction of the average magnetron frequency contribution $\bar{\nu}_-(t_{accu})$ from the free cyclotron frequency $\nu_c(t_{accu})$. Here, $\bar{U}(t, t_{accu})$ is the trapping voltage at the specific measurement time t averaged for t_{accu} . Two scenarios over a certain measurement time have to be determined. These scenarios are covering a fast and a slow voltage fluctuation. The fast voltage fluctuations do have minor influence, due to an averaged voltage fluctuation, which is balanced over a full measurement period, thus, these fluctuations are by far less critical. The slow trapping voltage fluctuations which are introduced by the term $\bar{U}(t, t_{accu})$, have to be minimized, due to their strong effect to increase the angular spot distribution by their average phase time t_{phase} to:

$$\delta\Phi = \frac{1}{t_{phase}} \sqrt{\int_0^{t_{phase}} (\Phi - \bar{\Phi})^2 dt}, \quad (4.30)$$

the phases Φ and the average phase $\bar{\Phi}$ ¹⁶ are determined by:

$$\Phi = \frac{t_{accu}}{2d^2 B} \cdot \bar{U}(t, t_{accu}) + const, \quad (4.31)$$

and

$$\bar{\Phi} = \frac{1}{t_{phase}} \int_0^{t_{phase}} \Phi dt = \frac{t_{accu}}{t_{phase}} \cdot \frac{1}{2d^2 B} \cdot \int_0^{t_{phase}} \bar{U}(t, t_{accu}) dt + const. \quad (4.32)$$

Including the Eqs. (4.31) and (4.32) into Eq. (4.30) results to:

$$\delta\Phi_{slow} = \frac{1}{2d^2 B} \cdot \frac{t_{accu}}{t_{phase}} \sqrt{\int_0^{t_{phase}} \left(\bar{U}(t, t_{accu}) - \frac{1}{t_{phase}} \cdot \int_0^{t_{phase}} \bar{U}(t', t_{accu}) dt' \right)^2 dt}. \quad (4.33)$$

Thus, the resolving power becomes hampered by the slow voltage drift, if the spot phase shift $\delta\Phi_{slow}$ exceeds the spatial distribution of the projected phase spot given by Eq. (4.11):

$$\arcsin \left(\frac{\Delta r_{det}}{r_{det}} \right) < \delta\Phi_{slow}. \quad (4.34)$$

¹⁶The phase shift is valid for both radial motions and will be therefore here omitted.

Re-writing the Eq. (4.34) leads to the following dependency:

$$f(t_{\text{phase}}) < r_{\text{det}} \cdot t_{\text{accu}}. \quad (4.35)$$

With this description it becomes clear that smearing of phase positions is defined by the excitation radius in correlation with the accumulation time t_{accu} . A reduction of the radius would thus simultaneously reduce the spatial smearing of the spot distribution, while at the same time reducing the resolution and accuracy counterproductively.

In order to give an overview of the correlation of the daily trap voltage fluctuations in comparison to the magnetron frequency and the temperature variations in respect with the reduced cyclotron fluctuations, the following figure Fig. 4.18 is presented:

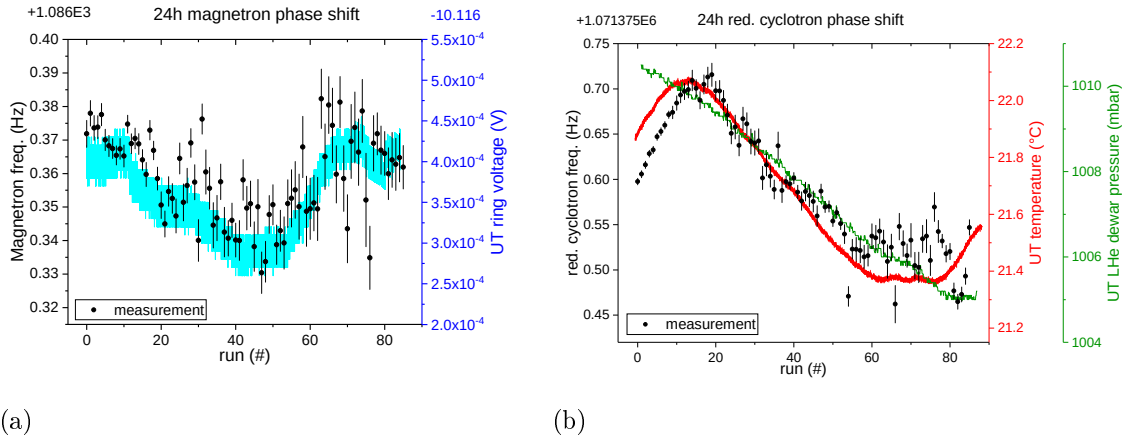


Figure 4.18: Illustrated are the magnetron frequency fluctuations (a)(left ordinate) recorded over 24 h at ISOLTRAP. These fluctuations are in comparison with the relative ring segment trapping voltage $\bar{U}(t, t_{\text{accu}})$ (a)(right ordinate)(cyan). The phase-accumulation time was $t_{\text{accu}} = 0.6$ s, where the ions were excited to a radius of $\rho_- = 0.6$ mm. In (b) a 24 h fluctuation of the reduced cyclotron frequency is compared to the precision Penning trap bore temperature (red) and the helium pressure (green) fluctuations. See text for details. The error bars are larger at the end due to a count rate problem, i. e. smaller statistics, at the last 20 measurements.

In Fig. 4.18(a) the daily voltage fluctuation¹⁷ $\bar{U}(t, t_{\text{accu}})$ in comparison with the magnetron frequency fluctuation is recorded. Where in (b) the daily fluctuation of the reduced cyclotron frequency is visualized. Both data sets are recorded in the direct PI-ICR-measurement scheme Section 4.1.2 in order to be able to distinguish the external influences from each other. With an excitation radius of $\rho_- = 0.6$ mm and a phase accumulation time of $t_{\text{accu}} = 0.6$ s the daily magnetron frequency shift does not exceed $\Delta\nu_- < 50$ mHz. Therefore, the phase shift of the phase spot due to the voltage fluctuation is about $\delta\Phi_{\text{slow}} = 18^\circ$ at maximum (Eq. (4.33)). Higher

¹⁷For this measurement a Keithley 2001 (7.5 digits) multimeter has been used.

voltage stability could be achieved with a more stable power supply, additionally, a temperature-stabilized housing that covers the power supply to maintain a more consistent temperature environment. In Fig. 4.18 (b) the reduced cyclotron frequency doesn't exceed a shift of $\Delta\nu_+ < 250$ mHz, respectively, $\delta\Phi_c = 70^\circ$, representing the temperature fluctuation (red) as the biggest influencing factor, respectively, the magnetic field fluctuation. Even small temperature fluctuations of $\Delta T = 0.05$ °C become noticeable in a frequency fluctuation of about $\Delta\nu_+ = 30$ mHz, as can be seen in Fig. 4.18 (b) between the measurement run 60 to 80. However, the liquid helium pressure fluctuation of the precision Penning trap dewar had minor influence on the cyclotron frequency (Fig. 4.18 (b) (green)). Other influencing factors, are the transport voltages from the preparation Penning trap towards the precision Penning trap. These fluctuations change the injection position, thus the initial magnetron radius and with it the starting position of the ν_+ excitation, which increases the systematic uncertainty, due to an admixture of radial motions. Further tests of the transport simulations and measurements must be carried out to determine whether the transport can be further stabilized. The following section examines how the voltage and temperature fluctuations described above influence the frequency determination during a measurement.

Non-simultaneous measurement of the reference and the final phase

Measuring the position of the reference phase (pattern 1), as described in Fig. 4.3 (a), is not sensitive to fluctuations of the radial frequency. This statement is only true as long as the duration of the dipole-excitation pulse, which defines the reference phase, is instantly, therefore, the excitation pulse's amplitude becomes infinite. For a non-instantaneous excitation time T_{ex} , the final position may vary after each excitation due to voltage instabilities, as explained in Section 4.4.5. Looking at the corresponding radial frequency shift between two measurements, a value of $\Delta\nu_{shift}$ can be obtained. According to the fact that the frequency is subject of the motion's phase, the angular position changes $\Delta\Phi_{shift}$ with respect to the frequency shift as in the following:

$$\Delta\Phi_{shift} = 2\pi \cdot T_{ex} \cdot \Delta\nu. \quad (4.36)$$

The uncertainty thus arises to:

$$\delta\nu = \frac{T_{ex}}{t_{accu}} \cdot \Delta\nu. \quad (4.37)$$

The daily fluctuation at ISOLTRAP, as mentioned in the previous subsection Section 4.4.5, of the radial motion induced by trapping voltage fluctuations does not exceed $\Delta\nu_U < 50$ mHz. The pulse duration of the magnetron excitation is set to three cycles, which leads to $T_{ex,mag} = 2.759$ ms. A more extensive test to reduce the magnetron excitation duration to one magnetron cycle $T_{ex,mag} = 0.919$ ms is still pending. In case of the dipole-excitation pulse of the reduced cyclotron frequency, the amplitude to convert with shorter pulses is too small. The issue at ISOLTRAP

are the FAN IN/ FAN OUT modules¹⁸, which prevent to apply amplitudes higher than 2 V to the ring segments. In case of the reduced cyclotron frequency the current excitation time is therefore, $T_{ex,red} = 217 \mu\text{s}$. The voltage fluctuation for example in case of the magnetron frequency results to an uncertainty of:

$$\frac{\delta\nu_U}{[\text{Hz}]} < \frac{T_{ex}}{t_{accu}} \cdot \frac{\Delta\nu_U}{[\text{Hz}]} \approx \frac{1.4 \cdot 10^{-4}[\text{s}]}{t_{accu}}, \quad (4.38)$$

where t_{accu} is given in seconds. The shift has a minor influence with the current precision, but could be lowered to be $\delta\nu_{mag,U} = 4.6 \cdot 10^{-5} \text{ Hz}$. In all other dipole excitation cases with the reduced cyclotron frequency, the current uncertainty reaches $\delta\nu_{red,U} = 1.1 \cdot 10^{-5} \text{ Hz}$. By use of the direct measurement scheme of the magnetron and the reduced cyclotron motion, to determine the free cyclotron frequency, the trapping voltage fluctuations cancel each other and thus it seems that the free cyclotron frequency is not affected by trapping voltage fluctuations. This statement is, however, only true if both measurements could be done simultaneously or the voltage fluctuations are on average the same in both measurement cycles, as discussed in Section 4.4.5. Taking into account the trapping voltage fluctuation and the quadrupole conversion time of $T_{conv} = 1.4 \text{ ms}$, the influence on the cyclotron frequency results to $\delta\nu_{c,U}/\nu_c < 6.5 \cdot 10^{-11}$, which could be neglected with the current systematic uncertainties.

Since the voltage fluctuations have now been discussed, the magnetic field fluctuations and their effects on the determination of the free cyclotron frequency ν_c must be checked. A typical daily fluctuation of the cyclotron frequency at ISOLTRAP is $\Delta\nu_{c,B} < 250 \text{ mHz}$, dominated by the reduced cyclotron frequency fluctuation. The magnetic field stability, affected by pressure and temperature fluctuations was discussed in Section 2.2.3 and was determined to be $\delta B/\delta t \cdot 1/B \sim 6.4 \cdot 10^{-10}/h$ [Ata16]. The influencing effects are demonstrated in Fig. 4.18 (b). Nevertheless, an active temperature stabilization could stabilize the magnetic field, which might reduce the daily fluctuations of the cyclotron frequency to a lower limit of a few tens of mHz. Without the temperature stabilization, the current influence on the frequency due to the quadrupole conversion time $T_{conv} = 1.4 \text{ ms}$, is determined to be:

$$\frac{\delta\nu_{c,B}}{[\text{Hz}]} < \frac{T_{conv}}{t_{accu}} \cdot \frac{\Delta\nu_{c,B}}{[\text{Hz}]} \approx \frac{3.5 \cdot 10^{-4}[\text{s}]}{t_{accu}}. \quad (4.39)$$

Taking into account the fluctuations determined in Eq. (4.39) and by assuming an accumulation time of $t_{accu} = 1 \text{ s}$, the relative cyclotron frequency shift due to magnetic field instabilities does not exceed $\delta\nu_{c,B}/\nu_c < 3.3 \cdot 10^{-10}$ for a singly charged $^{85}\text{Rb}^+$ ion. Thus, this influence with the current precision at ISOLTRAP can be neglected, but has to be addressed for long term measurements, if the independent measurement scheme Section 4.1.1 would be used.

¹⁸The FAN IN/ FAN OUT (FIFO) module uses one input signal and multiplies the signal to several outputs.

4.4.6 Inclinations and their impact on the frequencies

The effects of a tilted detector by an angle of δ_{tilt} and a linear displacement s with respect to the trap axis, respectively, a tilt of the magnetic field symmetry axis related to the electric field axis Θ , as shown in Fig. 4.19, is discussed in the following.

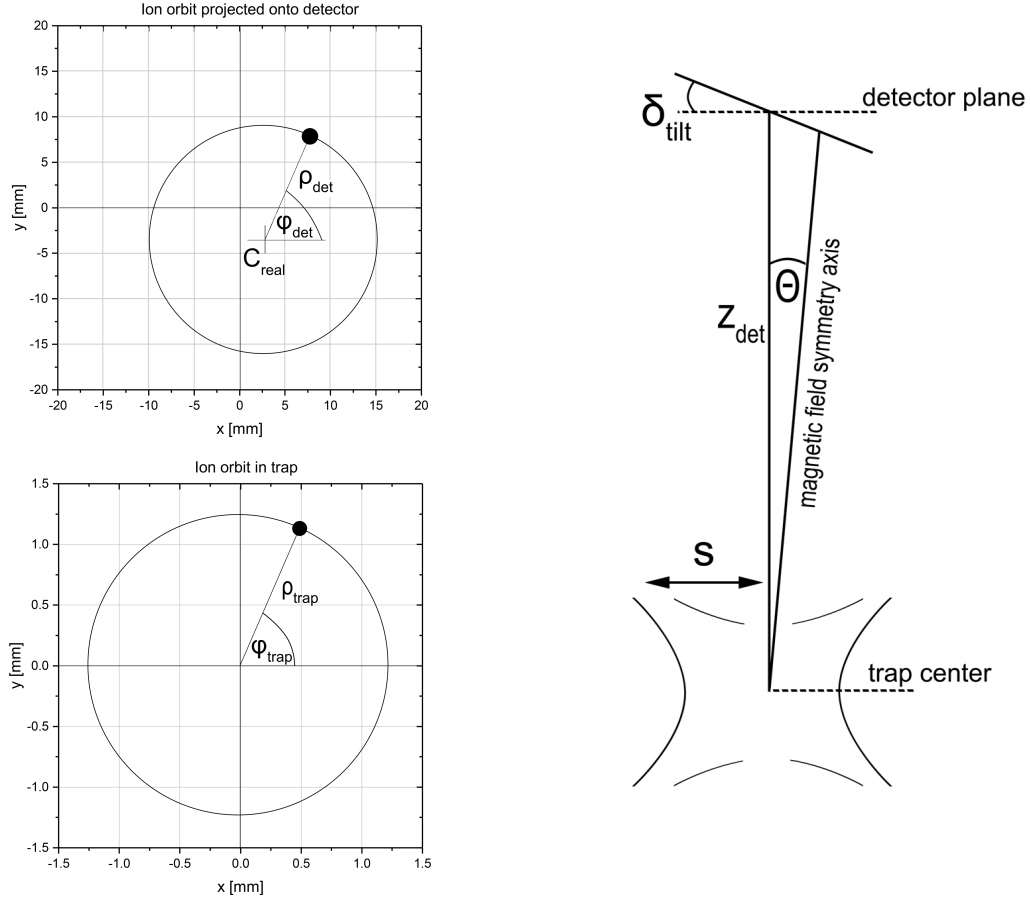


Figure 4.19: In picture (a), the ion orbit in the trap and the ion path projected onto the detector are visualized. The caused distortion by the magnetic field tilt Θ with respect to the electric field axis, as well as the detector misalignment δ_{tilt} is clarified in (b), as well as the linear displacement s .

Ion magnification factor

As previously mentioned, the angular displacement of the magnetic field symmetry axis with respect to the trap axis at ISOLTRAP should not exceed $\Theta = 0.08^\circ$, due to the alignment intervention in 2013 with an electron gun [Ata16]. To introduce the coordinates of the ion in the trap $(\rho_{trap}, \varphi_{trap})$ with their linear dependence to the corresponding projection on the detector $(\rho_{det}, \varphi_{det})$ with its projection factor

P , can be written as:

$$(\rho_{det}, \varphi_{det}) = P \cdot (\rho_{trap}, \varphi_{trap}). \quad (4.40)$$

The magnification represented by the projection factor can be written in a simple analytical form as long as the ion moves at low speed along the magnetic field lines in an electrical field-free region. This could be in principle achieved by applying the same voltage to all drift tube elements D1-D6, as introduced in Table 4.3. In case of ISOLTRAP's construction, these electric field-free boundary conditions can not be fulfilled. Further details which deal with these issues and how these errors become noticeable will be discussed at the end of this subsection.

In order to introduce the behavior of the magnification factor of the ions, a non-perturbative electric field-free region is first considered, in which the ions follow only the magnetic axial field lines. The magnification can then be described as:

$$\begin{aligned} \rho_{det} &= P_B \cdot \rho_{trap}, \\ \varphi_{det} &= \varphi_{trap}. \end{aligned} \quad (4.41)$$

Here, P_B denotes the magnification factor without an additional electric field gradient. By use of Gauss's flux law, the magnification P_B is given to:

$$P_B = \frac{\rho_{det}}{\rho_{trap}} = \sqrt{\frac{B_z(\rho_{trap}, 0)}{B_z(\rho_{det}, z_{det})}}, \quad (4.42)$$

where the axial component of the magnetic field of the precision Penning trap at radius ρ_{trap} is determined to $B_z(\rho_{trap}, 0) = 5.9$ T and for the detector axial position $z_{det} = 913$ mm at radius ρ_{det} with $B_z(\rho_{det}, z_{det}) = 0.016$ T. The magnetic field strength at both positions were measured during the maintenance in 2013, as described in [Ata16]. This results in the magnetic field driven total magnification factor for ISOLTRAP:

$$P_B \approx 20. \quad (4.43)$$

The magnification factor can be considered constant for $\rho_{det} < 20$ mm, since the magnetic field lines within the trap can be considered parallel over a small trap volume. This result can also be obtained by an excitation of a well-known radius of $\rho_{trap} = 0.4$ mm, in comparison to the obtained projected radius on the detector, e.g. $\rho_{det} = 8$ mm. The following Fig. 4.20 is a demonstration of the electrical and magnetic field magnification factor P at ISOLTRAP, whereby all drift electrode voltages are varied simultaneously.

The small observed beating results from the different acceleration voltages, respectively, ion velocities, probing different magnetic field trajectories, on which the ion spirals along towards the detector¹⁹. A main point becomes immediately clear when comparing the magnification factor $P \approx 23$, obtained in Fig. 4.20, with the maximum possible magnetron radius of $\rho_- = 1.2$ mm. Due to the high magnification factor, not all attainable radial motion phases can be projected onto the detector

¹⁹The higher the potential, the larger the velocity, the larger the period of the beating becomes.

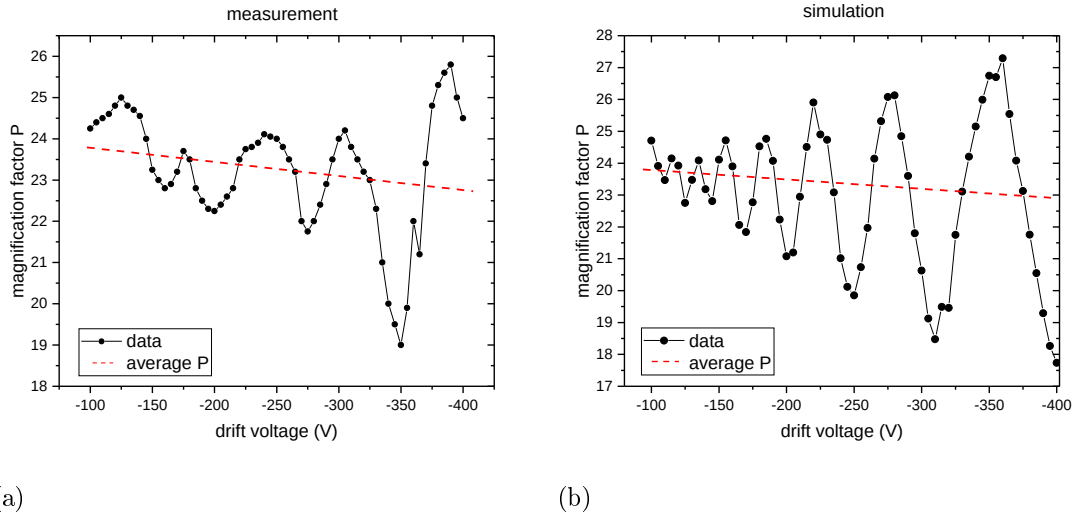


Figure 4.20: The obtained magnification factor P , measured at ISOLTRAP with respect to different drift electrode voltages (a), in comparison to a SIMION simulation on the contrary side (b). The used magnetron radius of the ion orbit in the precision Penning trap and the simulation was $\rho_- = 0.4$ mm. Shown are the data points, as well as the average magnification factor highlighted by the dashed red line as expected.

with the aid of the current structure. In order to avoid this limiting effect, two different procedures could be followed to meet the necessary projection conditions. a) The detector could be lowered of about 25 cm towards the trap center, where the magnetic field strength would increase to $B_{lower} = 0.06$ T or b) the voltages of the drift electrodes could be set to $U_{drift} = -1500$ V to achieve an electric field gradient free projection with the current detector position. Unfortunately both scenarios cannot be achieved at ISOLTRAP. On the one hand in scenario a) the presently used position-sensitive detector (DLD40) (Section 4.3.1), does not function properly in an environment with such a high magnetic field strength. On the other hand a further lowering of the detector is not possible due to ISOLTRAP's construction. The second statement b) does not hold as well, because of the distance between the trap ejection electrodes and the drift elements, which causes an electrical discharge for voltages higher than $U_{drift} > -400$ V. In order to meet the requirements nevertheless, a *SIMION* simulation with a simplex algorithm that changes the drift voltages by taking into account several starting parameters was accomplished. In the initial conditions of this simplex algorithm [Kar17] also the inclinations of drift elements 3&4, and 5&6 to each other were considered. While an additional parameter dealt with the condition for maximizing the achievable spatial ion-distribution-to-radius ratio, as described in Section 4.2.1. The resulting magnification factor sufficient for the current ISOLTRAP setup has been determined to be $P_{sim} = P = 5.5$ by simulation. This value was experimentally confirmed, as shown in Fig. 4.4, where the ToF-ICR magnetron radius ρ_- was compared to the projected magnetron radius by use of the PI-ICR technique. The obtained voltages have already been introduced in Table 4.3.

To achieve the real position of the orbit-image center C_{real} , the previously introduced angular Θ and linear s displacement have to be considered. In case of the angular displacement the following formula meets the condition [Eli14]:

$$\begin{aligned} C_{real,\Theta} &= z_{det} \cdot \Theta \cdot \frac{2\pi}{360}, \\ &\approx 16 \cdot \Theta \approx 0.8 \cdot P_B \cdot \Theta, \end{aligned} \quad (4.44)$$

where the linear displacement yields to:

$$C_{real,s} = (P_B - 1) \cdot s. \quad (4.45)$$

The angular displacement Θ is given in degrees, where z_{det} , as well as the linear displacement s in mm. Thus, the shift of the image center has no impact on the frequency determination, whereas the image shape admittedly does. If the angular displacement Θ is considered as an inclination of the radial ion motion and the detector plane with respect to the magnetic field trajectories, then the orbit image remains circular. The orbit-image center of that projected orbital affected by the displacement is shifted by the value of $C_{real,\Theta} = 1.3$ mm, relative to the symmetry axis of the the trap electrodes. In case of a small linear displacement of the trap symmetry axis with respect to the homogeneous magnetic field center volume, in which the magnetic field lines can be regarded as parallel, the following formula describes the displacement as follows:

$$\rho_{det} = P \left(s \cdot \cos(\varphi_{trap}) \pm \sqrt{\rho_{trap}^2 - s^2 \cdot \sin^2(\varphi_{trap})} \right). \quad (4.46)$$

Accordingly to Eq. (4.46), a small linear displacement has no effect on the ion orbit in the trap and its projection. Due to the fact that the magnification factor P can be considered constant, while the ion follows only another magnetic field line that is shifted by the amount of the linear displacement of the factor s . A small note beside: A linear displacement of the detector relative to the symmetry axis of the trap center can still be helpful, referring to the maximum magnification factor at ISOLTRAP of $P = 23$. This magnification factor can be used to achieve the highest resolving power Section 4.2.2, i. e. a higher accuracy, with the disadvantage that not all magnetron motion phases can be projected at once onto the position-sensitive detector's active plane.

Tilted detector

The shifts described in the previous subsection did not affect the orbital image of the ion. Within this subsection, the tilt δ_{tilt} of the detector which certainly has an effect, will be surveyed. Any inclination of the detector symmetry with respect to the trap electrodes symmetry axis will result into an ellipticity of the orbit image. The distortion factor results from $\epsilon = 1/\cos(\delta_{tilt})$, as introduced in Fig. 4.19 (b).

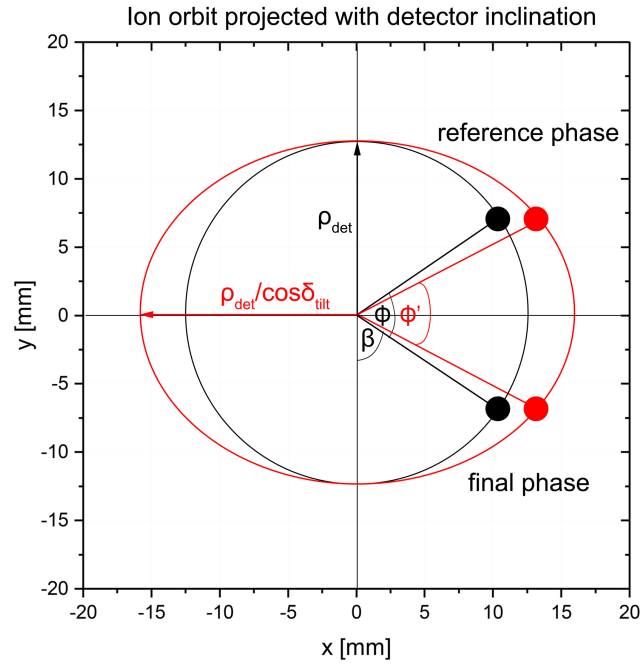


Figure 4.21: Each ion orbit projection onto a detector with a certain inclination leads to an elliptical distortion, which is marked in red. Compared to a perfectly aligned detector as shown in black. Measuring with a non-perfectly aligned detector would lead to different phase angles between the reference and final phase if they do not overlap, respectively, to incorrect frequency determination. [Eli14].

This elliptical orbit projection causes a change of the length of the projected circular magnetron radius $\rho_{-,trap}$, while probing the whole magnetron-motion's phase, as shown in Fig. 4.21:

These changes in the projected radius will cause a radial frequency error if the reference and final phase do not overlap [Eli14]. Therefore, the maximum phase shifts have to be taken into account. In case of the magnetron frequency determination the phase shift exceeds $\Delta\Phi_U = \pm 9^\circ$ at ISOLTRAP within 24 h, where in case of the cyclotron frequencies the phase shifted about $\Delta\Phi_c = \pm 35^\circ$ (Section 4.4.5). To estimate the uncertainty introduced by the misalignment, the latter phase shift is used in the following:

$$\begin{aligned} \Delta\nu_{tilt} = \nu - \nu' &= \frac{\Phi - \Phi'}{2\pi t_{accu}} \\ &= \frac{\Phi - \left[\arctan\left(\frac{\tan(\beta+\Phi)}{\cos \delta_{tilt}}\right) - \arctan\left(\frac{\tan(\beta)}{\cos \delta_{tilt}}\right) \right]}{2\pi t_{accu}}. \end{aligned} \quad (4.47)$$

Here, the obtained radial frequency from the undistorted ν and distorted ν' orbit images, respectively, the angles Φ and Φ' are determined as shown in Fig. 4.21. The real final phase is thereby defined by the polar coordinate angle β . As mentioned before, in 2013 ISOLTRAP's precision Penning trap was aligned with a precision of

$\Theta = 0.08^\circ$. With the assumption for the maximum daily phase shift of $\Phi = 70^\circ$ and a maximum detector tilt of $\delta_{tilt} = 1^\circ$, ϵ will be smaller than 1.0003, therefore, the frequency shift results to:

$$\frac{\Delta\nu}{[Hz]} < \frac{6 \cdot 10^{-5}[s]}{t_{accu}}. \quad (4.48)$$

Taking into account the cyclotron frequency obtained, which was introduced in Table 2.1 as $\nu_c = 1.072$ MHz, for a singly charged $^{85}\text{Rb}^+$ ion in a $\mathbf{B} = 5.9$ T magnetic field, the shift of the free cyclotron frequency is $\Delta\nu_c < 5.6 \cdot 10^{-11} [s]/t_{accu}$. Thus, the impact of the uncertainty caused by the misalignment of the detector with respect to the trap axis or the electrical field symmetry axis with respect to the magnetic field axis within the current uncertainties can be ignored. However, if a higher precision is achieved, the detector can be aligned even better than $\delta_{tilt} < 1^\circ$, e. g. by the procedure described below.

An additional method to cross-check the ellipticity factor ϵ directly, instead using an electron gun, from which the whole trap would have to be replaced, is described in the following Fig. 4.22:

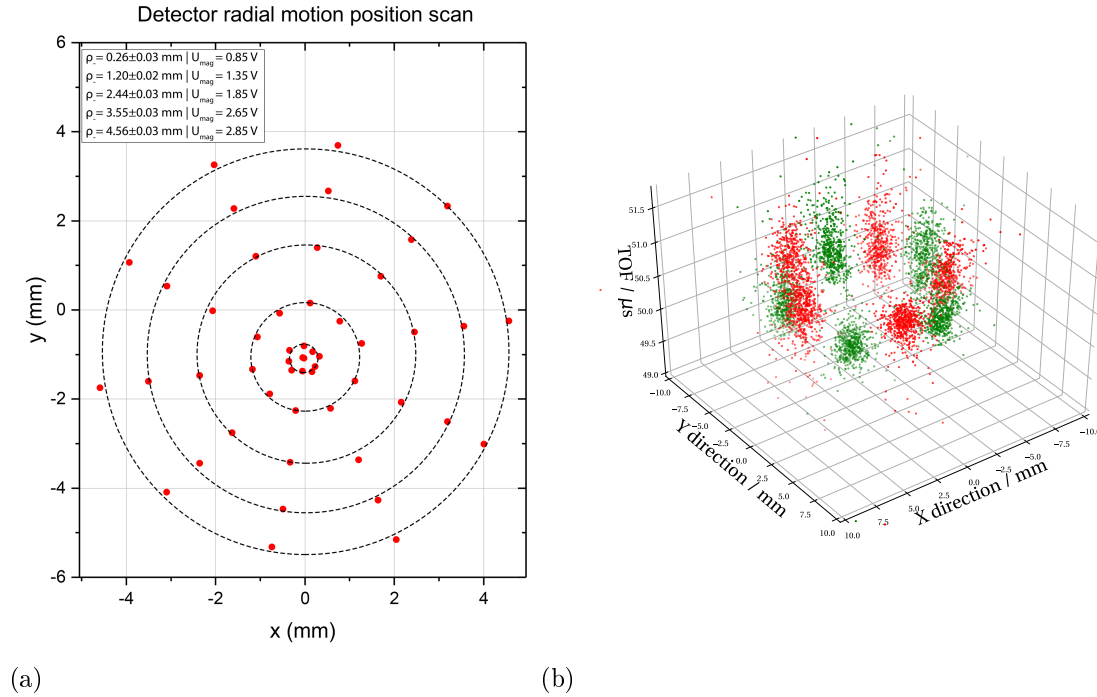


Figure 4.22: Figure (a) shows a method for estimating the inclination with the symmetrical spot distribution on the position-sensitive detector, where the two-dimensional ion projection with the ToF information as the third dimension is shown on the opposite side in (b). Both values can be compared and correlated to each other. The planar coordinates are determined by the position-sensitive detector, where the third dimension is spanned by the time-of-flight distribution of the arriving ions impinging on the MCP. For reasons of clarity, (b) does not show all ion distributions. See text below for more information.

The detector inclination δ_{tilt} can be directly determined, by measuring a set of ion ejections on the magnetron motion, along the magnetron-phase. For example ten ejection points at different magnetron radii to probe the projection plane x , y and their diagonal lines, as can be seen in Fig. 4.22 (a). To avoid misinterpretation, the time-of-flight information, as presented in Fig. 4.22 (b), can be used to build a correlation map for each ion spot distribution. With this additional information, an inclination of the detector in relation to the magnetic field, as well as the electrical field axis, would be perceptible in the form of different flight times or different radial positions, while a perfectly aligned detector would produce an even ToF distribution for each specific magnetron radius and without position ellipticity. It should be mentioned that the ToF values of the individual measurements can only be used for comparison with each other in relation to the inclination, if the axial motion is not disregarded as a decisive disturbing factor, since this becomes noticeable as a strong ToF oscillation, as shown in Appendix A.5 and Appendix A.6. Thus, the magnetron phase scan should be performed in steps of the axial motion period so that the phase of the axial motion is locked.

The following example shows how well the adjustment can be carried out with the technique described above. Considering the average total flight time of e. g. $^{85}\text{Rb}^+$ ions at ISOLTRAP with $t_{ToF} = 50.3 \mu\text{s}$ at a total flight length of $z_{det} = 913 \text{ mm}$, an ion velocity of $\mathbf{v}_{ion} = 18.2 \text{ mm}/\mu\text{s}$ results. With a projected magnetron radius of $\rho_{det} = 7 \text{ mm}$ and an additional inclination of the detector of, for example, $\delta_{tilt} = 1^\circ$ would result in a 0.12 mm shorter flight distance for the ions on one side of the detector. Taking into account the ion velocity, the ions would reach the detector at the side of the shorter flight distance $\Delta t_{ToF} = 7 \text{ ns}$ earlier, compared to a well aligned detector. This relatively large ToF difference can be resolved with the TDC8HP converter and its resolution of $t_{res} = 25 \text{ ps}$, as described in Section 4.3.2. With respect to this resolution and the flight time of $^{85}\text{Rb}^+$ at ISOLTRAP, the smallest noticeable change in distance is in the order of $\sim 1 \mu\text{m}$, thus, the alignment of the detector could be better than $\delta_{tilt} < 0.008^\circ$.

The described procedure can also be simulated in the *SIMION* environment and correlated with the results obtained to determine the ion velocity or axial energy of the ion. Further tests have to be done to prove this type of direct determination of the ellipticity, before it can be considered as a reliable technique.

It should be noted that the linear displacement at ISOLTRAP in the x -direction of the position-sensitive detector plane doesn't exceed $\Delta x_s < -2 \text{ mm}$, where in case of the y -axis with respect to the axis center the linear displacement is smaller than $\Delta y_s < -0.5 \text{ mm}$. These values are obtained by applying the same voltage to each drift electrode to ensure that the ions fly unperturbed in an electric field free region. By use of the simulated voltage set 4.3 to achieve the best spatial-distribution-to-radius ratio Section 4.2.1, the displaced center $C_{trap,s,\Theta}$ moved almost to the center of the active detector plane. This effect is most likely due to the focusing effect of the ejection voltage with respect to the first two drift elements. Thus, it will not influence the projection of the phase spots. Further tests have to be carried out

in order to exclude a displacement of the preferred ion path by a misaligned focus element. With the construction of ISOLTRAP, it is foreseen to adjust the trap tower containing the electrostatic optics as well as the trap itself with respect to the magnetic field's symmetry axis. The whole tower is aligned within the so called SUMA-tube (superconducting magnet tube). The SUMA-tube is fixed in place by using a cardanic support structure. With this structure it is possible to change the trap's center symmetry axis in relation to the magnetic field symmetry axis, whereby the fully equipped trap tower is placed in the magnet vessel. This advantage leads to the possibility that the position-sensitive detector can be used as a live position adjustment monitor to align the whole trap tower at once. The adjustment will take place in an iterative process between a position measurement and the next alignment step to minimize the angular Θ and the linear s displacement. This method can also be used for the preparation Penning trap and its cardanic support to minimize the initial magnetron radius that occurs when the preparation Penning trap and the precision Penning trap are not aligned to the same magnetic symmetry axis. Nevertheless, at ISOLTRAP the injection into the preparation Penning trap can be manipulated by steerer elements along the transfer path of the ions between both traps. This provides the possibility of a proper injection, respectively, a minimization of the initial magnetron motion. Further tests at ISOLTRAP to investigate the transport stability and its effects on the associated systematic influences are still pending.

4.4.7 Interconversion of radial motions and the shift effect of

$$\nu_+$$

As previously mentioned in the indirect Section 4.1.1 or the direct Section 4.1.2 pattern, the ions which orbit on the fast reduced cyclotron motion must be converted after a certain step into the slow magnetron motion to ensure that the ions experiencing the same trajectory after each ejection. The quadrupole conversion π -pulse was introduced in Section 2.2.4, it is the sideband excitation at $\nu_c = \nu_+ + \nu_-$. It was introduced that the time of the conversion, respectively, the phase $\Delta\Phi$, the ion accumulates over a certain time T_{conv} , is dependent from the strength of the conversion amplitude. A simultaneous conversion at a fixed point from one motion to another is therefore only possible with an infinite amplitude height. Technically, ISOLTRAP's construction is limited by the maximal amplitude $U_{quad} = 20$ Vpp²⁰ the used frequency generators (33510B Agilent) are able to provide. Another limiting factor is the FIFO modules used, which supply the ring segments of the precision Penning trap with the rf-signals of the frequency generators. By using these FIFO modules, the conversion signal is attenuated by a factor of three. Thus, the quadrupole conversion period lasts 1500 cycles, which corresponds to a conversion time of $T_{conv} = 1.4$ ms. The ions therefore experience several phases before they reach their endpoint. Under perfect conditions this endpoint would be the same after each conversion for the *reference* phase (pattern 1) and the *final* phase (pat-

²⁰The acronym *Vpp* describes the maximum voltage between peak-to-peak.

tern 2), respectively. As it has been clarified, the trap is subject of the temperature and thus, among other things, the accompanying side effects of voltage fluctuations Section 4.4.5 and magnetic field fluctuations Section 4.4.3. Due to these influences, both the *reference* and *final* phases are affected by these disturbances, leading to a phase drift over time. The radial solutions for each motion were introduced in Eq. (2.42). To visualize the ion motion's behavior during a quadrupole conversion, the radii,

$$\rho_{\pm}(t) = \sqrt{\frac{2\hbar}{m(\omega_+ - \omega_-)}} \cdot |\alpha_{\pm}|, \quad (4.49)$$

respectively, the complex amplitudes $\alpha_{\pm}(t) = |\alpha_{\pm}(t)| \cdot e^{\mp i\phi_{\pm}(t)}$ will be introduced to:

$$\begin{aligned} \alpha_{\pm}(t) = e^{\mp i(\omega_{\pm} + \delta_{dt}/2) \cdot t} \cdot [& (\cos\left(\frac{\omega_R t}{2}\right) \pm i \frac{\delta_{dt}}{\omega_R} \cdot \sin\left(\frac{\omega_R t}{2}\right) \cdot \alpha_{\pm}(0) \\ & - i \frac{2g}{\omega_R} \cdot \sin\left(\frac{\omega_R t}{2}\right) \cdot e^{\mp i\Phi_{rf}} \cdot \alpha_{\mp}(0), \end{aligned} \quad (4.50)$$

as explained in [Kre12].

In Eq. (2.42) the Rabi-frequency was introduced as $\omega_R = \sqrt{(2g)^2 + \delta_{dt}^2}$ describing the beating frequency of the quadrupole conversion, in relation to a detuned frequency $\delta_{dt} = 2\pi(\nu_{rf} - \nu_c)$. In the following, the perfect case without an initial magnetron radius is used at the beginning. The pure reduced cyclotron motion is fully converted into the magnetron motion, therefore only the start and end conditions can be considered in the following. The start phase has the complex amplitude $\alpha_+(0)$, respectively, motion phase $\Phi_+(0)$, where the amplitude after the conversion is $\alpha_-(T_{ex})$ and the phase $\Phi_-(T_{ex})$, which can be given to [Eli14]:

$$\begin{aligned} |\alpha_-(T_{ex})| \cdot e^{+i\Phi_-(T_{ex})} = & \frac{2g}{\omega_R} \cdot \sin\left(\frac{\omega_R T_{ex}}{2}\right) \cdot |\alpha_+(0)| \\ & \cdot e^{+i(\omega_- + \frac{\delta_{dt}}{2})T_{ex} + \Phi_{rf} - \Phi_+(0) + \frac{3\pi}{2} + 2\pi k}. \end{aligned} \quad (4.51)$$

The final amplitude, as well as the final phase after the conversion results to:

$$\begin{aligned} |\alpha_-(T_{ex})| = & \frac{2g}{\omega_R} \cdot \sin\left(\frac{\omega_R T_{ex}}{2}\right) \cdot |\alpha_+(0)|, \\ \Phi_-(T_{ex}) = & \left(\omega_- + \frac{\delta_{dt}}{2}\right) T_{ex} + \Phi_{rf} - \Phi_+(0) + \frac{3\pi}{2} + 2\pi k. \end{aligned} \quad (4.52)$$

The parameter k is in integer. A full conversion without detuning $\delta_{dt} = 0$ leads therefore to $|\alpha_+(0)| = |\alpha_-(T_{ex})|$ and with it to:

$$g \cdot T_{ex} = \frac{\pi}{2} + 2\pi p, \quad (4.53a)$$

$$\Phi_-(T_{ex}) = \omega_- \cdot T_{ex} + \Phi_{rf} - \Phi_+(0) + \frac{3\pi}{2} + 2\pi k. \quad (4.53b)$$

The value p is expressed in steps of integer. By this assumptions the difference of the *final* phase and the *reference* phase of the cyclotron motion ($\Phi_+^f - \Phi_+^r$) will

be converted into the magnetron corresponding *final* phase and *reference* phase ($\Phi_-^f - \Phi_-^r$), which results to:

$$\Phi_-^f - \Phi_-^r = -(\Phi_+^f - \Phi_+^r). \quad (4.54)$$

This preserves the *final* phase and the *reference* phase angle through full quadrupole conversion at ν_c . By use of Eq. (4.54) it gets clear that the ion's cyclotron motion have opposite angular directions, as the magnetron motion's image spot. As discussed in Section 4.4.5, the temporal instabilities of the trapping voltages are able to shift the magnetron frequency of the *final* phase and *reference* phase by a certain amount of $\delta\nu_-$. This shift can be used to express the error of the cyclotron phase to $\delta\Phi_+ = 2\pi \cdot \delta\nu_{-ex}$. Under this assumption, the relative error of the cyclotron frequency evolves to:

$$\frac{\delta\nu_+}{\nu_+} = \frac{\delta\nu_- \cdot T_{ex}}{\nu_+ \cdot t_{accu}}. \quad (4.55)$$

Assuming the typical daily variation at ISOLTRAP's magnetron frequency of about $\delta\nu_- = 50$ mHz for a singly charged ion with a reduced cyclotron frequency $\nu_+ = 1.072$ MHz, in the case of ^{85}Rb and a conversion time of $T_{ex} = 2.759$ ms resulting to:

$$\frac{\delta\nu_+}{\nu_+} = \frac{6.5 \cdot 10^{-11}[\text{s}]}{t_{accu}}. \quad (4.56)$$

The introduced error by this fluctuation can be neglected in case of the magnetron motion. In case of the daily magnetic field fluctuations, which are subject of the free cyclotron frequency, the error $\delta\nu_c$ which results from the conversion can be obtained as in the following:

$$\frac{\delta\nu_+}{\nu_+} = \frac{\delta\nu_c \cdot T_{ex}}{2 \cdot \nu_+ \cdot t_{accu}}. \quad (4.57)$$

Taking into account the conversion time of $T_{ex} = 1.4$ ms and the daily free cyclotron frequency drift of $\delta\nu_c = 250$ mHz due to the magnetic field fluctuations, the relative uncertainty of ν_c results to:

$$\frac{\delta\nu_+}{\nu_+} = \frac{1.6 \cdot 10^{-10}[\text{s}]}{t_{accu}}. \quad (4.58)$$

Reflecting only the raising uncertainties introduced by the phase shift in comparison with the current achievable uncertainty and by use of the direct measurement scheme, both the magnetron and cyclotron frequency drift can be neglected with regard to the conversion phases.

So far only the perfect scenario with a pure reduced cyclotron motion has been reflected. Taking an initial magnetron amplitude $\alpha_-(0) = |\alpha_-(0)| \cdot e^{+i\cdot\Phi_-(0)}$ admixture into account introduces an additional shift, which leads to an error in the determination of the free cyclotron frequency. By considering a full conversion as described in Eq. (4.52), the coupling factor g with an additional initial magnetron

motion can be described by [Kre12]:

$$g \cdot T_{ex} = \frac{\pi}{2} + 2\pi p + \frac{1}{2} \cdot \arctan \left[2 \sin(\Phi) \cdot \frac{S}{S^2 - 1} \right]. \quad (4.59)$$

Here, $\Phi = \Phi_{rf} - \Phi_-(0) - \Phi_+(0)$, where $S = |\alpha_-(0)|/|\alpha_+(0)|$ determines the ratio between the initial magnetron and the reduced cyclotron motion amplitudes. The error which results for a certain accumulation time t_{accu} for the determination of the cyclotron-motion phase is then given by $\Delta\Phi = (\Phi_-^f - \Phi_-^r) + (\Phi_+^f - \Phi_+^r)$:

$$\begin{aligned} \Delta\Phi = 2\pi - \arctan 2 \left[\frac{1 - k(f) \cdot \sin(\Phi^r + \omega_c t_{accu})}{k(f) \cdot \cos(\Phi^r + \omega_c t_{accu})} \right] \\ + \arctan 2 \left[\frac{1 - k(r) \cdot \sin(\Phi^r)}{k(r) \cdot \cos(\Phi^r)} \right], \end{aligned} \quad (4.60)$$

both angular wave numbers $k(f)$ and $k(r)$ are given by:

$$\begin{aligned} k(f) &= \frac{2 \sin(\Phi^r + \omega_c t_{accu}) \cdot \frac{S^2}{S^2 - 1}}{1 + \sqrt{1 + 4 \sin^2(\Phi^r + \omega_c t_{accu}) \cdot \frac{S^2}{(S^2 - 1)^2}}}, \\ k(r) &= \frac{2 \sin(\Phi^r) \cdot \frac{S^2}{S^2 - 1}}{1 + \sqrt{1 + 4 \sin^2(\Phi^r) \cdot \frac{S^2}{(S^2 - 1)^2}}}. \end{aligned} \quad (4.61)$$

As a consequence of Eq. (4.60), the phase conversion error becomes zero when the phase-accumulation time t_{accu} evolves in multiple of half the period of the free cyclotron motion $\omega_c t_{accu} = \pi n$. Assuming a realistic achievable case at ISOLTRAP, where the reduced cyclotron radius is $\rho_+ = 0.6$ mm and the initial magnetron radius is $\rho_- = 0.025$ mm, thus, the radius ratio results to $S = 0.05$. If the radius ratio between the magnetron and the reduced cyclotron motion exceeds larger values than $S > 0.1$ or a higher precision is necessary than $\Delta\nu_c = 10^{-9}$, the phase-accumulation t_{accu} has to be a multiple of half the period of the free cyclotron frequency to preserve an overlap of the *reference* and the *final* phase spot. This can be achieved only to a certain level, due to temporal magnetic field instabilities Section 4.4.3. Additionally, the to be measured masses are only to a certain extend known or predicted with a big uncertainty of about 0.5 - 1 MeV/c² [NL18]. The daily free cyclotron frequency fluctuation of an ⁸⁵Rb⁺ ion at ISOLTRAP measures $\Delta\nu_c = 250$ mHz, respectively, an uncertainty of $2.3 \cdot 10^{-7}$. Thus, the cyclotron frequency has to be known to at least $1 \cdot 10^{-6}$ so that the corresponding uncertainty covers the last 2π -phase evolution on the position sensitive detector within the given phase accumulation time $\omega_c t_{accu}$. Therefore, the to be measured frequency has to be known to at least 100 mHz, determined, e.g. with the standard ToF-ICR technique.

Admixture of motions

Finally, it should be noted that the initial axial motion has to be as small as possible, as demonstrated in Section 4.4.1 to avoid a smearing of the spatial-distribution spot. A larger ion ToF distribution projected with a large magnification factor P onto the detector will end up in an elliptical spot, because of a tail of slower ions. This effect is caused by the ions which spiral along the deviating magnetic field lines. In the following Fig. 4.23, a real measurement (a-c) and a simulation (d-f) are shown in a scenario under extreme conditions:

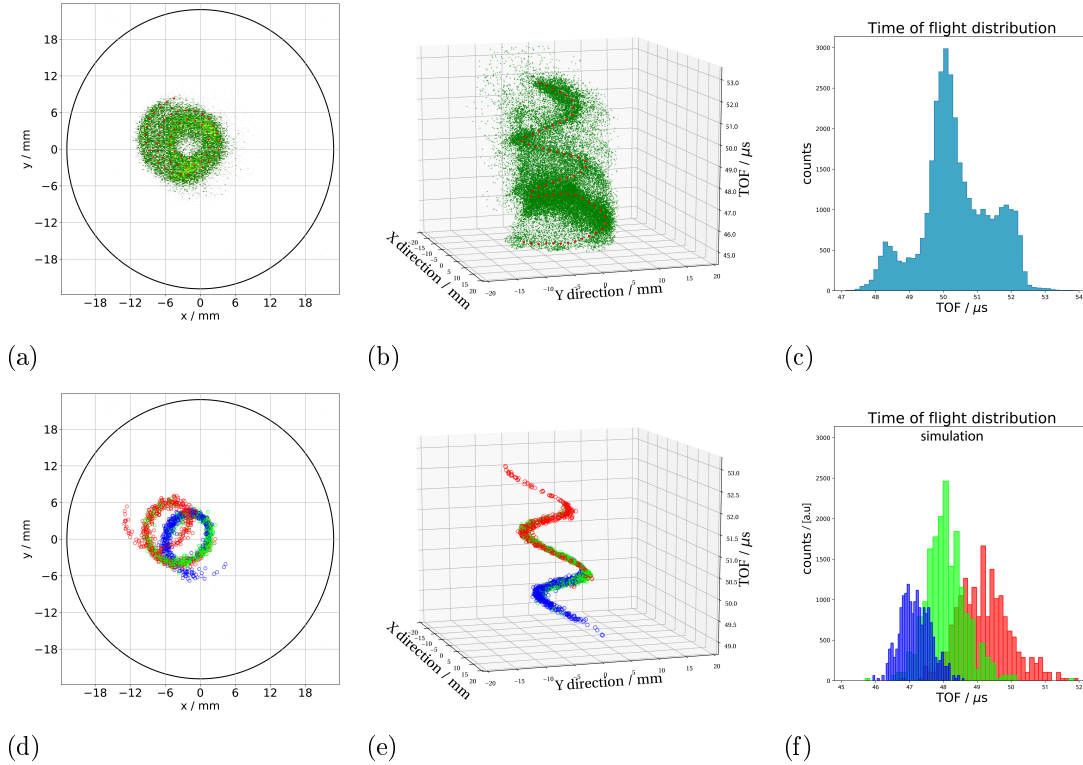


Figure 4.23: Projection of ions with a large axial motion and an admixture of the reduced cyclotron and magnetron motion. These starting conditions result in an elliptical spot projected towards the 2D-plane (x&y-axis), as shown in picture (a). In (b) the time-of-flight information (z-axis) in combination with the 2D-plane information result into a 3D projection. By this procedure, the ions which spiral along magnetic field lines can be visualized. In figure (c) the time-of-flight distribution of the measured ions is shown. In comparison to the measurement, a simulation as shown in (d) - (f) will help to draw new conclusions about the processes of the ions in the trap and on their way to the detector.

In Fig. 4.23, the ion motions behavior are demonstrated with a remaining axial motion, an initial magnetron motion, as well as a quadrupole conversion at the wrong conversion amplitude, respectively, conversion time T_{conv} . These extreme conditions help to elucidate possible causes of distorted, spatially distributed spots. For the measurement in Fig. 4.23, a twice as large axial motion than the minimal achievable axial motion at ISOLTRAP was used. To demonstrate the ion spiraling

behavior along the magnetic field lines an admixture of the fast reduced cyclotron motion and the slow magnetron motion was created. The projected spiral makes itself felt as an elliptical projected spot in the 2D-plane Fig. 4.23 (a). In combination of the position-sensitive information and the time of flight, a 3-dimensional picture can be generated, which represents the real ion behavior (b). It is important to say that the spiral does not immediately indicate the local behavior of the ion within the trap. Nevertheless, the time information might be used in comparison with a simulation, as shown in Appendix A.4, e.g. to extract the axial energy of the ions in the trap. This procedure needs to be examined more closely and should only be mentioned here for the sake of completeness.

4.4.8 Search for additional accumulation time shifts

A crucial limiting parameter to determine the ion's motions precisely is the accumulation time t_{accu} , defining the time the ion orbits within a specific motion. At the beginning of the implementation phase of the PI-ICR technique at ISOLTRAP, as discussed in Section 4.3.3, the FPGA card with its resolution of $t_{\text{res}} = 25$ ns was already compared to the internal time jitter of the frequency generator. To exclude further uncertainties of the accumulation time, more tests were done, which will be described in the following.

The internal time precision between the trigger acknowledgment of CH1 (e.g. dipole excitation ν_+) with respect to CH2 (e.g. π -pulse) in the used Agilent 33510B frequency generators is smaller than $\Delta t_{\text{delay}} < 200$ ps²¹. However, this time is therefore a crucial factor, if it is not a fixed value and if this value is not taken into account as an additional time for the accumulation time t_{accu} . The total accumulation time which has to be used for the evaluation of the frequency, as described in Eq. (4.2), is the total accumulation time of $t_{\text{accu-tot}} = t_{\text{accu}} + \Delta t_{\text{delay}}$. Other influences that can change the accumulation time are explained subsequently. These influences have to be splitted into two groups, the constant offsets Δt_{offset} and the dynamic changes Δt_{dyn} . A constant offset is not a crucial factor, if all signal lines experience the same offset.

- Constant t_{accu} offsets: The accumulation time offset Δt_{offset} is composed by, e.g. the used FIFO (Philips 744). At ISOLTRAP the currently used module delays the signal of about $\Delta t_{\text{FIFO}} \sim 10$ ns. Additionally, the FIFO distributor module is connected with a galvanic decoupling box to the ring electrodes. The ring electrodes are connected with the decoupling box by 12 m LEMO cables, which accumulate to a delay of $\Delta t_{\text{cable}} \sim 60$ ns each. The total delay of about $\Delta t_{\text{offset}} = \Delta t_{\text{FIFO}} + \Delta t_{\text{cable}} + \Delta t_{\text{other}} = 70$ ns per output, is not a crucial issue as long as this delay is a constant offset for all distributed frequencies.

²¹As described in the Agilent 33510 manual.

- Dynamic t_{accu} changes: Dynamic changes in accumulation time are not a decisive factor, unless the dynamic changes are changes outside of a measurement cycle or the changes are rather small compared to the achieved systematic uncertainties. These dynamic changes can then be treated as an additional constant offset during a measurement. One example for a dynamic change is the ambient temperature or vibrating cables in vicinity of a strong magnetic field. These influences can change, among others, the impedance and with it the signal delay of each cable. To minimize these effects, the cables are firmly connected to each other so that the signals cannot be distorted with respect to each other.

In order to check whether each cable supplying ISOLTRAP's ring electrodes has the same length, the time domain reflectometer (TDR) method was used. This technique uses the principle of a pulsed function generator and an oscilloscope. With the oscilloscope the reflected and superimposed pulse signal's propagation time is determined as the cable length. Within the resolving power, limited by the used function generators rising time of about 8.4 ns, all four signal paths can be considered equal in length. To be able to validate the real cable length the resolution has to be increased. For example by use of a TDR with a simple set of Schmitt triggers (e. g. 74AC14) as a function generator for cable length measurements in combination with an oscilloscope would result in a < 2 ns rise time. A much more precise method would be the frequency domain reflectometer (FDR). The FDR uses a swept RF signal instead of TDR-DC pulses. In FDR technique, the pulse generator applies different frequencies to the cable to be measured. The response of the system is now queried on the same line with an oscilloscope, but in the time (distance) domain which is the result after the frequency is inverse Fast Fourier Transformed $f(t) = \sum_{n=-\infty}^{\infty} F_n \cdot e^{jn\omega_{rf}t}$ into the time domain.

The explained delays within this subsection at ISOLTRAP with respect to the timing are most of the time constant signal offsets. These offsets do not influence the total accumulation time $t_{\text{accu-tot}}$. The remaining offset between the internal trigger delay of CH1 and CH2 of each function generator, which independently controls the P1 or P2 patterns, must be considered as the additional accumulation time offset if a higher precision should be achieved. Within the current uncertainties, this offset of $t_{\text{accu-tot}} < 200$ ps, which affects the uncertainty in the order of $\delta\nu_c/\nu_c < 5 \cdot 10^{-10}$, can be neglected.

4.4.9 Summary of possible uncertainties at ISOLTRAP

The uncertainties mentioned in this section, which occur at the cyclotron frequency, are summarized in Table 4.4 in comparison to the determined values at SHIPTRAP/GSI [Eli14]:

Reason of systematic cyclotron frequency shifts: $\delta\nu_c/\nu_c$	@ ISOLTRAP		@ SHIPTRAP
	$^{85}\text{Rb}^+$	$^{133}\text{Cs}^+$	$^{133}\text{Cs}^+$
Inclination/linear displacement	$< 5.6 \cdot 10^{-11}$	$< 8 \cdot 10^{-11}$	$< 10^{-11}$
Non-simultaneous measurement of <i>ref</i> - and <i>final</i> -phase	$< 1.6 \cdot 10^{-10}$	$< 2 \cdot 10^{-10}$	$\approx 10^{-10}$
Shifts caused during excitation and conversion times:			
Trapping voltage temporal instabilities	$< 6.5 \cdot 10^{-11}$	$< 2.1 \cdot 10^{-10}$	$\approx 10^{-10}$
Magnetic field temporal instabilities	$< 3.3 \cdot 10^{-10}$	$< 5.4 \cdot 10^{-10}$	$< 5.5 \cdot 10^{-11}$
Frequency detuning $\delta = \omega_{rf} - \omega_c$	$< 10^{-10}$	$< 10^{-10}$	$\approx 10^{-11}$
Minimal initial magnetron motion	$< 10^{-10}$	$< 10^{-10}$	$< 5 \cdot 10^{-10}$

Table 4.4: Typical systematic shifts of the free cyclotron frequency $\delta\nu_c/\nu_c$ for the PI-ICR detection technique at ISOLTRAP with $^{85}\text{Rb}^+$ and $^{133}\text{Cs}^+$ in comparison to SHIPTRAP with $^{133}\text{Cs}^+$ [Eli14].

In comparison with ISOLTRAP to SHIPTRAP, the temperature-stabilized magnet from SHIPTRAP is noticeable in the magnetic field stability. While in the case of the magnetron motion, a higher voltage stability can be achieved with ISOLTRAP. An advantage of SHIPTRAP over ISOLTRAP is the fact that the ions are transported on axis from the preparation Penning trap into the precision Penning trap, as both traps are located within the same magnet, which minimizes the initial magnetron motion. Where, in contrast, ISOLTRAP is in favor of long-time measurements due to the separation of both traps, whereby with ISOLTRAP a 2.5 times longer accumulation time can be achieved than with SHIPTRAP [Eli14], due to less residual gas interaction in the precision Penning trap (Fig. 4.17). Both experiments are able to handle the current achievable uncertainty of $\delta\nu_c/\nu_c > 10^{-9}$ by use of the PI-ICR technique. Taking into account the advantages and disadvantages of each individual will help to further improve the technique for future measurements.

Chapter 5

Mass measurements with PI-ICR

This section introduces the first determination of the excitation energy of an isomeric state using the PI-ICR technique at ISOLTRAP. The studied cases are $^{123g,123m}\text{Cd}$ and $^{127g,127m}\text{Cd}$.

The target material for the attempted cadmium beam time was chosen to be 50 g/cm² uranium-carbide, heated to about 2000°C in combination with a tungsten cylinder as neutron converter, as described in Chapter 3. The cadmium PI-ICR measurements evaluated here were conducted with the old detection arrangement behind the Penning trap, as described in Section 4.3.3. Thus, the thoroughly studied mass-independent error of 8×10^{-9} ([Kel03]) was used for the determination of the following masses. Since ISOLTRAP has been realigned in 2013, the re-estimated relative mass-dependent error of $3.5 \times 10^{-10}/u$, as determined in Section 2.2.7, has been used. In addition, to account for the average non-linear magnetic field drift, as already described in Section 2.2.3, the value $\Delta B/t = 6.35 \times 10^{-11} \text{ T/min}$ was used for the following mass determination.

5.1 $^{123,127}\text{Cd}$ ground states and their isomeric masses

Within this section, a brief summary of the masses measured with the PI-ICR technique in 2016 is presented. The results, taking into account further measurements from 2017 with higher accuracy, will be published elsewhere.

The importance of the precise known cadmium masses in vicinity of the magic nucleus ^{130}Cd and the importance for the r -process¹ as an anchor point is well described in the publication of D. Atanasov *et. al* [AW15]. This anchor point serves, among other things, to describe the equilibrium between, the β -decay, the neutron capture and neutron evaporation, where latter two are highly dependent on the binding energy. An example that illustrates the importance of this fact is the description of the origin of heavy elements produced in, e.g. type-II supernovae as well as from compact object binary mergers [Jus15]. At the time [AW15], the mass separation

¹The rapid neutron capture process [AGT07].

of the ground state (g.s.) and the isomeric state (i.s) in the measured isotope of $^{129g,m}\text{Cd}$ was not possible, see Appendix A.7. The ToF-ICR technique was not able to separate both states, due to half-life limitation of $T_{1/2} = 151.5$ (5.7) ms for the ground-state and $T_{1/2} = 147$ (3) ms for the isomeric case [Aud17]. The short half-life limited the excitation time, respectively, the mass resolving power, as described in Section 2.2.7. The existence of an isomeric state in ^{129}Cd was demonstrated in [Yor13], therefore, the mass measurement published in [AW15], was hampered by an admixture of the g.s. and i.s., during the beam time in 2014. This leads to an inaccurate determined mass, due to an admixture of the excited state. Thus, to a shifted cyclotron frequency of the ground state, as described in Section 2.2.3. To determine this remaining unknown mass of the ground state of ^{129}Cd , the PI-ICR technique was used for the first time at ISOLTRAP in an online run in 2016, achieving a mass separation of $^{123,127}\text{Cd}$, as presented in the following. The second successful measurement campaign with PI-ICR in 2017 which led to a successful record of the masses in ^{129}Cd of both states, in combination with the excitation trend of the spin $11/2^-$ isomeric state in $^{123,127,129}\text{Cd}$, respectively, the corresponding neutron-hole state ($\nu 1h_{11/2}$), will be published elsewhere.

5.1.1 Measurement

As in the case of the standard ToF-ICR technique, the frequencies are determined in PI-ICR for each measured isotope one after the other. With a sandwich structure of three consecutive measurements, ordered as $\nu_{c,ref}|\nu_{c,IoI}|\nu_{c,ref}$, the frequency ratio $r = (\nu_{c,ref}/\nu_{c,IoI})$ can be obtained, as described in Section 2.2.7. For each PI-ICR measurement a ToF-ICR measurement was recorded in advance to obtain a rough estimation of the free cyclotron frequency ν_c . With this information the accumulation time t_{accu} leads to the correct number of full rounds in each eigenmotion n_+ and n_- (Eqs. (4.1) and (4.2)). Before each measurement, a center point with no cyclotron excitation or π -pulse, but with a centering magnetron pulse, was recorded (Fig. 4.1). The direct measurement scheme, as introduced in Section 4.1.2, determines in pattern 1 the magnetron motion and thus acts as the *reference* phase. Whereas in pattern 2, the *final* phase is determined in form of the modified cyclotron frequency. In pattern 2, the ground and the isomeric states were separated within an accumulation time of $t_{accu} = 300$ & 600 ms in case of $^{123g,m}\text{Cd}$ and $t_{accu} = 150, 200$ & 300 ms for the measurement of $^{127g,m}\text{Cd}$. To determine the frequency more accurately, as previously introduced in Section 4.4.7, the phase spots either in P1 or P2 have to overlap each other. This can be only fulfilled in a sandwich measurement, where in pattern 2 either the g.s. overlaps with the pattern 1 phase spot or the i.s.. To overlap either the g.s. or the i.s., the accumulation time was adjusted for P1 and P2 in the same manner, so that either the g.s. or the i.s. overlapped with the pattern 1 spot. For an overview of the resulting differences, whether the points overlap or not, see [Kar17]. In total 10 PI-ICR measurements were obtained in case of $^{123g,m}\text{Cd}$ and 12 measurements were recorded for $^{127g,m}\text{Cd}$.

The phase spot distribution exceeded $2\Delta r = 6$ mm (2016), due to the strong defocus introduced by the old drift tube configuration, as can be seen in case of ^{127}Cd Fig. 5.1:

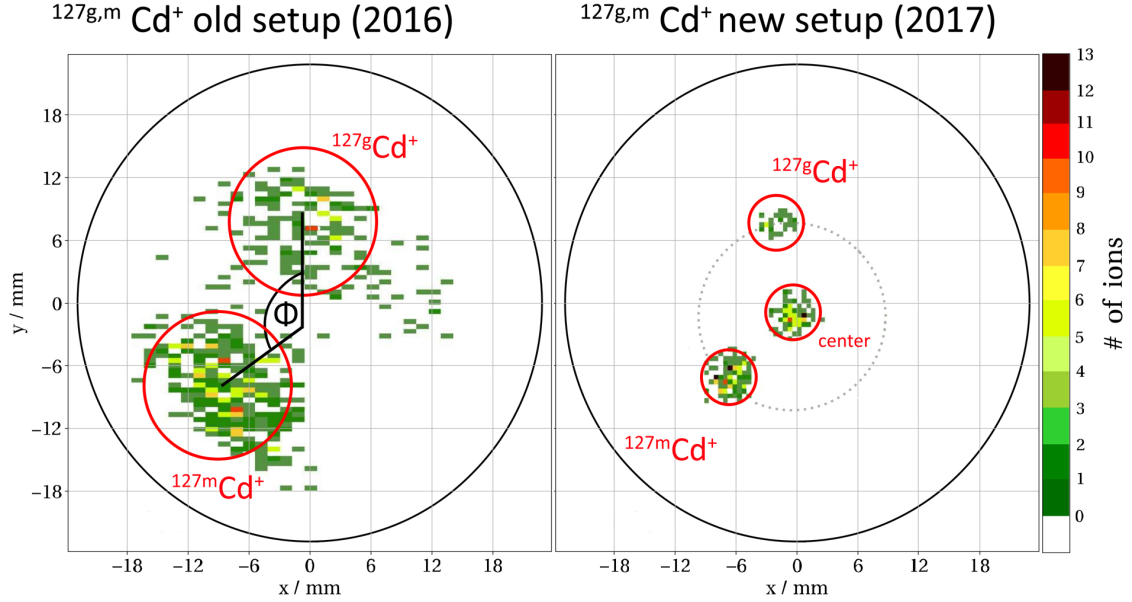


Figure 5.1: The separation of the $11/2^-$ isomeric-state from the $3/2^+$ ground-state of $^{127}\text{Cd}^+$ with an accumulation time of $t_{\text{accu}} = 150$ ms is presented. Highlighted are the ion count-rates, which impinged onto the 2D-plane of the position-sensitive MCP detector [Kar17]. In comparison, the old voltage set on the left hand side (2016) and the measurement with improved new voltages on the right hand side (2017) are shown.

The improvement between the settings in 2016 and 2017 is presented for the sake of completeness. The evaluation of the recorded data in the measurement campaign in 2017 will be published elsewhere. The achieved resolving power in 2016 of $\mathfrak{R}_{\text{PI-ICR}} = 10^5$ - 10^6 was sufficient to resolve and determine the mass of both states in case of $^{123,127}\text{Cd}$, as shown in the following results.

5.1.2 Results

The diffuse spots with a spatial distribution of $2\Delta r = 6$ mm (2016) hampered the mass determination only slightly, as can be seen in comparison with the obtained mass results in the following Table 5.1:

The masses of $^{123,127}\text{Cd}$ obtained in 2016 represent the first PI-ICR online measurements at ISOLTRAP, next to a measurement of $^{199,201}\text{Bi}$. The unpublished results of $^{127g,127m}\text{Cd}$ agree very well within one standard deviation with the new results from the TITAN/Triumf facility located in Vancouver/Canada [Las17]. The relatively large uncertainties of the PI-ICR measurement method compared to the measurements at TITAN, which used the standard ToF-ICR technique, are mainly

Table 5.1: Measured frequency ratios, as well as the experimentally determined mass-excess ME_{exp} values of $^{123g,m}\text{Cd}$ and $^{127g,m}\text{Cd}$. The measured masses were obtained in a sandwiched structure by use of $^{133}\text{Cs}^+$ ($ME(^{133}\text{Cs}) = -88070.931 (0.008)$ keV [Wan17]) as reference mass. Compared are the obtained mass-excess results to the AME, and in the case of ^{127}Cd with the TITAN/Triumf measurement.

Ions	$r = (\nu_{c,ref}/\nu_{c,IoI})$	ME_{exp} (keV)	ME_{AME} (keV)	ΔME_{AME} (keV)	ME_{Titan} (keV)	ΔME_{Titan} (keV)
$^{123g}\text{Cd}^+$	0.9248443036 (420)	-77415.17 (5.20)	-77414.20 (2.70)	0.97 (5.86)		
$^{123m}\text{Cd}^+$	0.9248455002 (411)	-77267.03 (5.09)	-77271.20 (4.83)	4.17 (7.02)		
$^{127g}\text{Cd}^+$	0.955011174 (113)	-68728.94 (13.93)	-68491 (13)	237.94 (19.05)	-68743.4 (5.6)	15 (15)
$^{127m}\text{Cd}^+$	0.9550133358 (910)	-68461.37 (11.27)	-68491 (13)	29.63 (17.21)	-68460.1 (4.7)	1.3 (12.2)

due to the larger systematic uncertainties caused by the diffuse spots in 2016, as shown in Fig. 5.1. In addition, the biggest issue besides the defuse spots, encountered during the beam time, was the imprecise knowledge of the accumulation time t_{accu} . ISOLTRAP's FPGA pattern card managed the accumulation time with just a resolution of $t_{res} = 25$ ns, as discussed in Section 4.3.2. In order to be able to estimate the true accumulation time nevertheless, the accumulation time trigger was in parallel recorded with the FAST MCS6A TDC counter, which has a $t_{bin} = 100$ ps resolution at an input rate of 10 Gbit/s. The obtained accumulation time trigger generated Gaussian distributed events whose center value represents the true accumulation time length, as shown below:

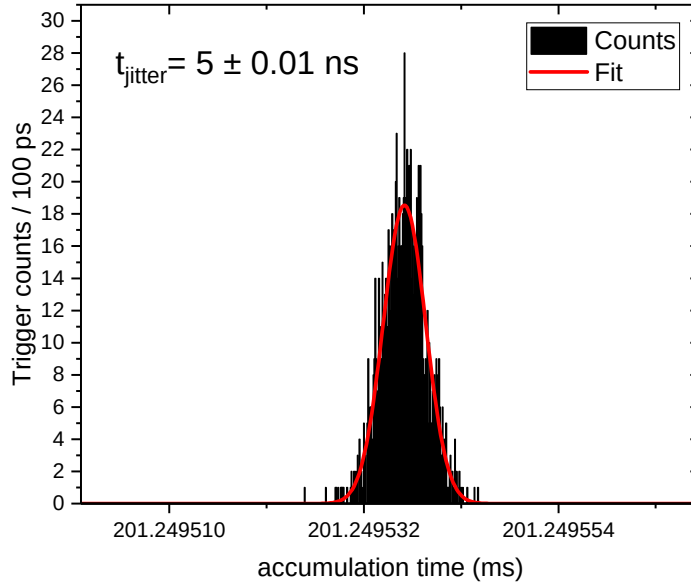


Figure 5.2: The recorded accumulation time trigger events for the FPGA card in case of an accumulation time $t_{accu} = 200$ ms.

The example shown in Fig. 5.2 results to a timing jitter of $t_{jitter} = 5 \pm 0.01$ ns. This results in an additional systematic uncertainty, e.g. as in case of ^{127}Cd , with an

accumulation time of $t_{\text{accu}} = 150$ ms, resulting in a contribution of $\delta m = 2$ keV (Section 4.1). The Gaussian distribution for the accumulation time t_{accu} was evaluated for each single measurement. During the cadmium beam time in 2016, the accumulation time trigger sent by the FPGA card determined whether the pattern 1 or the pattern 2 scheme was applied, as described in Section 4.1.2. This beam time illustrated the necessity to move to a frequency generator internal faster trigger solution with a more reliable accumulation time control, as described in Section 4.3.3.

5.1.3 Conclusion

For the first time at ISOLTRAP, the new installed PI-ICR technique was used to measure close lying isomers $\Delta E \sim 150\text{-}270$ keV and ground states, with short half-lives up to $T_{1/2} \sim 150$ ms. These mass measurements agree very well, within one standard deviation, to another facility which have used independently the standard ToF-ICR technique [Las17]. With this measurement it was demonstrated that the new technique can be used to determine and separate close lying isomers at ISOLTRAP. The obtained results will be compared to the new 2017 recorded data in a separate publication elsewhere. The here measured nuclei approach the $N = 82$ waiting point in the astrophysical r -process and are also of importance for the description of the nuclear mass surface as they lie near the doubly-magic nucleus ^{132}Sn , as mentioned in [AW15]. These information will help, amongst others to understand the r -process nucleosynthesis in a double neutron star merger ([Jus15; Pia17]).

Chapter 6

Summary and Outlook

A novel high-precision mass measurement technique was installed at ISOLTRAP within this thesis, which allows to reach currently a mass uncertainty as low as $\delta m/m \approx 10^{-9}$. The new Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique with its high resolving power allows to resolve close lying isomers (i.s.) and ground states (g.s.) compared to the standard Time-of-Flight Ion-Cyclotron-Resonance (ToF-ICR) techniques. Therefore, the technique could also be used for simultaneous half-life and hyperfine-structure measurements, as long as the isomeric- and ground states are produced in similar rate. It has been shown that the PI-ICR technique has an average resolution of 35 times better than the ToF-ICR technique. The measurement time by use of the PI-ICR technique to achieve the same accuracy as the standard ToF-ICR technique is of factor 25 improved. The ISOLTRAP setup with its four traps was firmly introduced, as well as the working principles of a Penning trap. With the introduction of the Penning traps, the standard ToF-ICR technique has been introduced. Subsequently, the explanation of the novel PI-ICR technique was discussed in detail. The two Penning trap measurement techniques used today at ISOLTRAP as standard mass spectrometry tools were compared, as summarized below. After a solid ground for the PI-ICR working principles were laid, the implementation steps through the different adaptation phases for the application of the new technique at ISOLTRAP were presented. Following the installation of the PI-ICR technique, the Penning trap tuning strategies for mass measurement for both ICR techniques were explained in detail. It was demonstrated that all tuning procedures can be done more precisely and faster with PI-ICR, rather than ToF-ICR. Only the real free cyclotron frequency determination has still to be done by the standard ToF-ICR technique with precision of at least $\delta m/m < 10^{-5}$ in advance, as a starting condition. The new tuning methods shown, among other things, included the harmonization of the electrostatic trapping potential, the magnetic field homogeneity adjustment, as well as the minimization of the axial and magnetron initial motions.

Beside the technical details of the various available techniques at ISOLTRAP, three different physics cases were presented, which are summarized in the following.

Starting with one of the ambitious ventures in physics, the determination of the neutrino mass, with which a mass measurement campaign puts its emphasis on the small electron capture Q_{EC} -values between the mass of the mother and the daughter nucleus. The smaller the remaining transfer energy, the more accurate the neutrino upper mass limit can be determined by other techniques, as e.g. Cryogenic Micro-Calorimetry (CMC). One of the candidates for determining the upper neutrino mass limit more precisely was the measured mass pair of ^{202}Pb - ^{202}Tl , which was published in the context of this work in [Wel17b]. The mass of ^{202}Tl was determined nine times more precise, leading to an improvement of the Q_{EC} -value precision of about factor three. This has shown that ^{202}Pb is not a bad candidate for its purpose, but it has also been shown that it might not be the most sensitive candidate, due to its high $Q_{\nu_e} = 23.5$ (4.3) keV value.

The second discussion on mass measurement discussed in this thesis deals with the area that deserves a strong focus of today's experimental and theoretical research on the nucleus structure of isotopes around the r -process waiting point of ^{78}Ni . Since the direct access of ^{78}Ni for mass measurements at ISOLDE is denied due to the 100-fold lower vapour pressure compared to copper, the mass surface of the $^{75-79}\text{Cu}$ isotopes one proton above the ^{78}Ni were studied instead and compared to large-scale shell model calculations (SM). The results are giving hints that ^{78}Ni has a doubly magic character. The obtained information by use of the SM calculations also show that to describe this region the protons and the neutrons have to occupy higher orbital levels across $Z = 28$ and $N = 50$ to minimize their energy. The results are published in PRL, as Editor's suggestion ([Wel17a]).

This thesis concludes with the evaluation of the first measured mass values of the resolved isomeric- and ground-state pairs $^{123,127}\text{Cd}$ obtained by the PI-ICR technique at ISOLTRAP in 2016. Both masses were compared with either the AME2016 or recent mass measurements from the TITAN group at Triumf/Canada to show that the PI-ICR technique is consistent in the uncertainties of one sigma to each other. With the new measurement campaign in 2017, the mass measurement of ^{127}Cd was confirmed with an even better control of the new PI-ICR technique. In addition, the mass of ^{129}Cd was resolved and accurately measured, which was not possible with the standard ToF-ICR technique in 2014 due to half-life limitations. The new obtained masses will be published elsewhere to demonstrate the impact to the waiting point of the magic ($N = 82$) ^{130}Cd nucleus, the r -process, as well as the increasing excitation energy trend of the $11/2^-$ isomeric states of $^{123,127,129}\text{Cd}$.

Outlook

The following outlook is a short summary of the ideas that could be addressed to further improve the used ToF-ICR and PI-ICR techniques at ISOLTRAP.

Several procedures to reduce the axial, the magnetron, as well as the reduced cyclotron motion were shown. Another attempt to further reduce the axial motion, in addition to the possibilities such as the cooling parameters in the preparation Penning trap and the correct capture time in the precision Penning trap, might be

the connection of a frequency generator between the upper and the lower end cap electrodes of the precision Penning trap using the same reduction principle, as in the case of the initial magnetron motion (Section 4.1). A subsequent step to be able to further reduce the energy of ion motions in the precision Penning trap might be a new crystal-resistive cooling technique that could be implemented in the ISOLTRAP system. Thus, two steps are necessary, first of all, the magnetron and the cyclotron motion must be efficiently reduced, therefore, a connection between the end cap and the ring electrode must be installed to be able to convert the radial motions into the axial motion [GXM93; Ita95]. The second step would be the installation of a crystal resonating circuit, which must be connected between both end caps of the precision Penning trap, to be able to pick up the axial stored energy by mirror charges, as it was shown for a crystal resonator that rested either in a room or cryogenic environment in [Kal11]. In order to increase the achievable mass range, a digital tunable crystal resistive cooling circuit must be designed, as suggested in [Kal11].

A calibration of the position-sensitive DLD40 delay-line detector is still pending. For this test, a calibration mask with a well-known pitch must be mounted in front of the MCP detector in order to achieve a higher spatial distribution reliability [Hon16]. The characterization of the detector can be achieved by using the ions available at ISOLTRAP or by installing an α calibration source in front of the detector.

Replacing several older devices is necessary, including the gate module, which decides whether to trigger the Pattern 1 or Pattern 2 frequency generator. The switching is currently carried out by an old frequency switch, as illustrated in Fig. 4.8. A new solid state switch should be implemented here to ensure a lower residual ripple (debounce) during switching and a lower attenuation of the TTL¹ trigger signal. Currently, a fan in/fan out module splits the excitation and conversion signals of the frequency generator into two-to-four parts for the ring segments, including the inversion of the phase of 180° . This process requires about 10 ns, which can be decisive if the two-to-four channels have a slight phase difference. In this case, the ring electrode with or without an amplifier can be connected directly to the output of the frequency generator with a capacitor in order to avoid a possible phase shift and to reduce the attenuation of the signal levels caused by such a module. Another advantage that supports the replacement of the fan in/fan out module, which follows the same approach, is a new amplifier for the frequency generators that provides a sufficiently high amplitude to further reduce the excitation and conversion times, as described in Section 4.4.7.

As a step towards an even higher precision for the PI-ICR technique, the randomization of the initial magnetron phase must be implemented in the control and data acquisition at ISOLTRAP. This allows to be able to have always the correct center of each spatially distributed spot during a longer measurement. Small injection fluctuations, which cause a different initial magnetron radius would be averaged and could thus help to reduce the systematic uncertainties. In order to enhance this step and to ensure the same starting conditions for all excitations, a magnetron de-excitation should be applied in future measurements before any further excitation

¹Transistor-Transistor-Logic

will be applied. This also includes the de-excitation pulse before each magnetron excitation, as required, e.g. in the independent measurement scheme Section 4.1.1. The scan routine to harmonize the electrical field in the precision Penning trap, as demonstrated in Section 4.4.3, can be optimized by a connection of the (PCI-6703) National Instruments controller card, which supplies the correction ring, the correction electrodes, as well as the correction tube voltages to the simplex algorithm at ISOLTRAP. With this simplex algorithm the trap can be automatically tuned to achieve the best harmonization of the precision Penning trap. In case of the magnetic field optimization, the z^2 room temperature shim coil current could be implemented in a computer controllable environment to automatically adjust the current to increase the stability of the system. These automatic tuning procedures would reduce the setup pre-tuning time, which has to be done before each beam time.

The idea presented to compare the simulated axial energies with the recorded three-dimensional figure (2D+ToF), as shown in Section 4.4.7 (Fig. 4.23), must be further tested in order to obtain the real ion energy. This might give rise to solutions for a new procedure to reduce the initial axial motion to an even smaller value.

A further approach to improve the Penning trap-assisted decay spectroscopy for ISOLTRAP would be an implementation of a re-acceleration beamline after the precision Penning trap. By means of PI-ICR mass separation, either one or the other state could then be ejected through a hole mask, which is followed by a 90° electrostatic bender and a re-acceleration cavity to implant finally the separated nuclide into a tape station². Thus, by use of a tape station a subsequent background free α -, β - and γ -spectroscopy of each state could be achieved. This might be important for the investigation of physical cases where both states, e.g., have the same energy regime, the same half-life, or are less produced and cannot be distinguished by the usual decay spectroscopy tools.

The physics cases summarized above can benefit from further developments on the ISOLDE side, e. g. different target materials, more efficient laser schemes and in the course of improving the precision of the PI-ICR technique.

In order to announce just a few examples, it should be proposed that further candidates from which the determination of the upper neutrino mass limit through a more precise determination of the Q_{EC} value could benefit are the pairs ^{193}Pt - ^{193}Ir and ^{157}Tb - ^{157}Dy [Wan17]. These masses could be addressed with the newly installed phase-imaging ion-cyclotron-resonance technique (Chapter 4), in a long term measurement campaign, to achieve the necessary uncertainty of a few $\delta m = 10$ eV for the Q_{EC} -value.

Due to the lower production rate and in consideration of the beam time, the access to the exact strength of the $N = 50$ neutron shell-closure of ^{79}Cu is still unknown. To determine the strength, the masses of $^{80,81}\text{Cu}$ must be measured to extract the S_{2n} , which is currently only extrapolated by SM ([Wel17a]). In order to address this

²A tape station is a type of cassette tape transport system on which nuclides can be implanted along the beam axis, which after a certain implantation time are transported to spectroscopy detectors located off the beam axis. This results in a smaller/free decay background spectroscopy.

field, a further improvement of the laser scheme and target material development must be achieved before the region of copper isotopes above the $N > 50$ shell-closer can be reached within a reasonable beam time length.

Now that ISOLTRAP has demonstrated its mass resolving power with the PI-ICR technique using $^{123,127,129}\text{Cd}$ further studies could be made, for example, the Ag isotopes region $^{118-126}\text{Ag}$ with many unknown isomeric states, for which a RILIS laser scheme with an ionization efficiency of $\leq 14\%$ [Fed95] at ISOLDE exists, whereby a sufficiently high yield is expected [Iso]. Another advantage of such a beam time would be to demonstrate the robustness of PI-ICR to a mass measurement of three different masses measured simultaneously, using e. g. ^{116}Ag , as a well-known benchmark mass, which has a double pair of isomers with sufficiently long half-lives.

Appendices

A.1 Simulated spatial ion distribution and its corresponding axial mean ToF

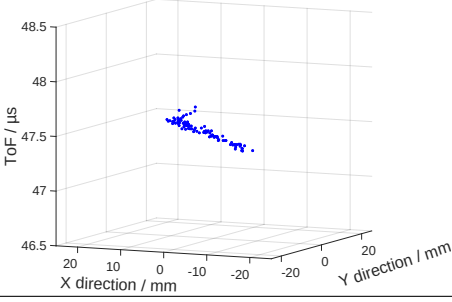
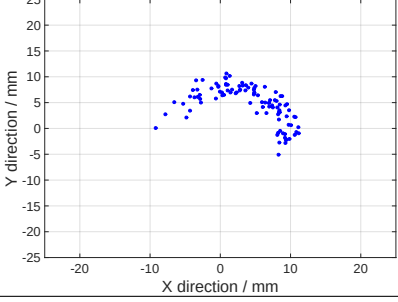
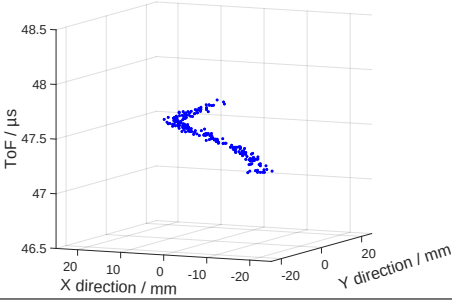
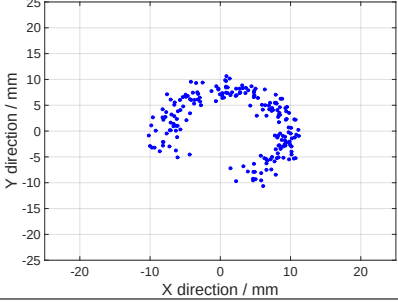
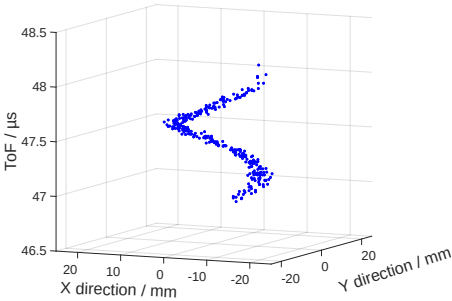
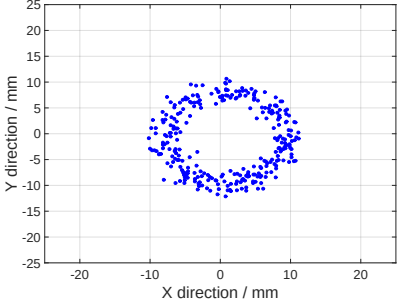
axial ToF	2D+ToF	spatial spread
500 ns $z = 1 \text{ mm}$		
750 ns $z = 3 \text{ mm}$		
1000 ns $z = 7 \text{ mm}$		

Table A.1: Simulated projection of ions of mass $A = 85$ and a charge of $q = +1$. Compared are different axial motion distributions in the Penning trap, which are superimposed by an admixture of the magnetron and the reduced cyclotron motion. It is clearly visible that the ions spiral along the magnetic field lines (2D+ToF), where the resulting projection is visible on the right hand side. The amplitude height z to each corresponding axial ion ToF distribution is highlighted below each value. Possible ways to manipulate the axial energy are mentioned in Section 4.4.1. For example, a solution is to change the ion capture time of the precision Penning trap, as shown in Appendix A.4.

A.2 Preparation and precision Penning trap transport voltages

drift electrodes	previous transport voltages [V]	new transport voltages [V]
Lens LT1	-380	-440
Deflector LT1x	-9	-8.5
Deflector LT1y	6.5	10
Transfer MCP3	-1000	-1000
Transfer LT	-1000	-1000
Lens LT2	-200	-320
Deflector LT2x	-0.25	-9.5
Deflector LT2y	9.2	6.4
Transfer MCP4	-1000	-1000
Transfer UT	-1000	-1000
Abschluss electrode	-1000	-1000
UT Inj. Transfer	-1000	-1000
Deflector UT1x	23.6	8
Deflector UT1y	20	6.2
Lens UT2	-240	-430
Deflector UT2x	(-3 to 5)	(1 to 2)
Deflector UT2y	(-2 to 9)	(1 to 2)
Lens UT3	-285	-510
Retardation 1	-810	-70
Retardation 2	-400	-25

Table A.2: The previous and the new, less mass-dependent voltages for the transfer electrodes between the preparation Penning trap (lower trap: LT) and the precision Penning trap (upper trap: UT) are shown. All used focusing and steering elements are floated with an additional $U_{float} = -1000\text{V}$.

A.3 Lower trap and upper trap magnetic field

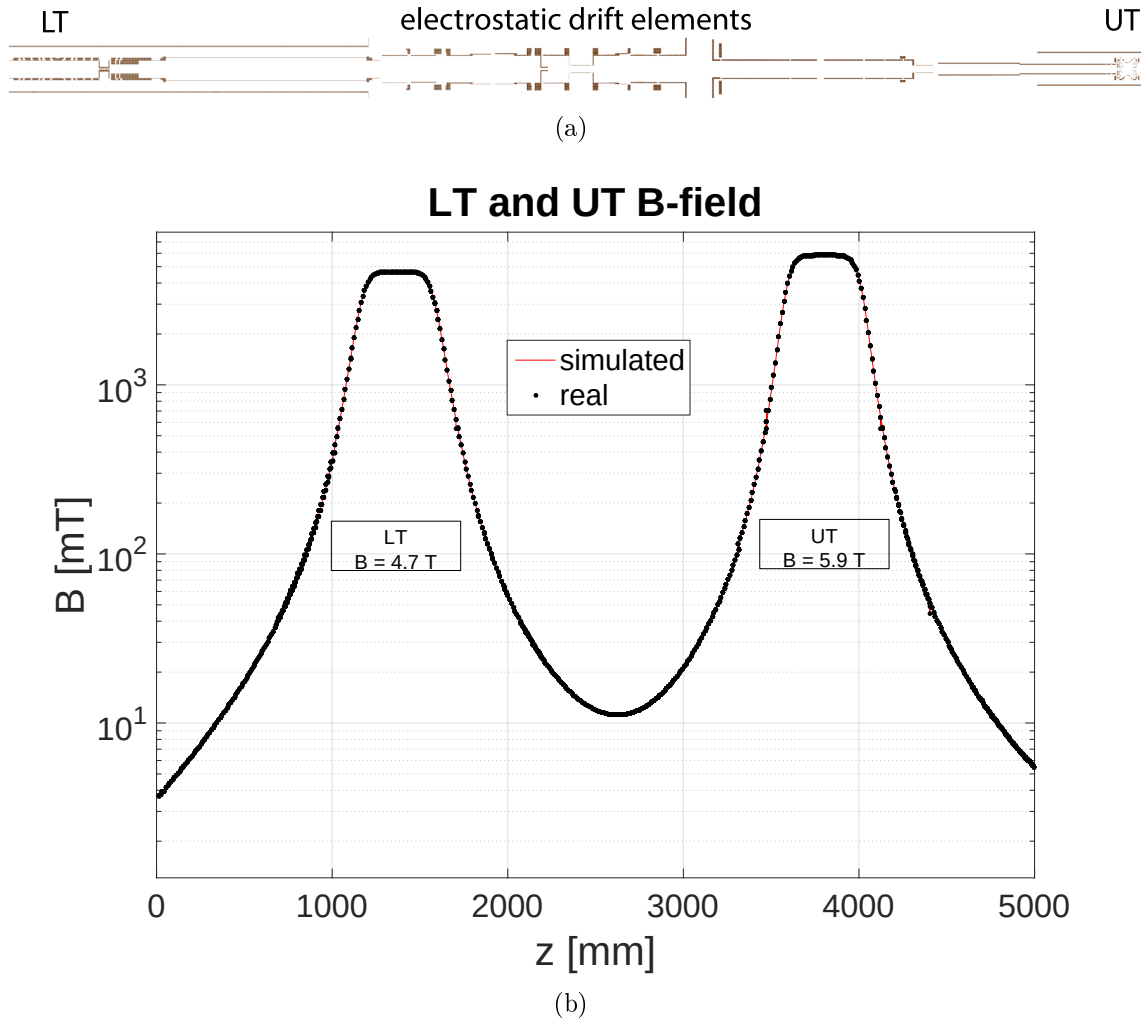


Figure A.3: The upper picture illustrates the LT, the UT and the geometry in SIMION. The lower picture represents the measured and by us of SIMION's Biot-Savard solver, simulated magnetic field gradient, of both traps at ISOLTRAP. The simulation environment presented here simulates the ion transfer from the LT into the UT. These settings were used to simulate the UT capture delay studies, as discussed in Section 4.4.3.

A.4 Upper trap capture delay and axial deflection

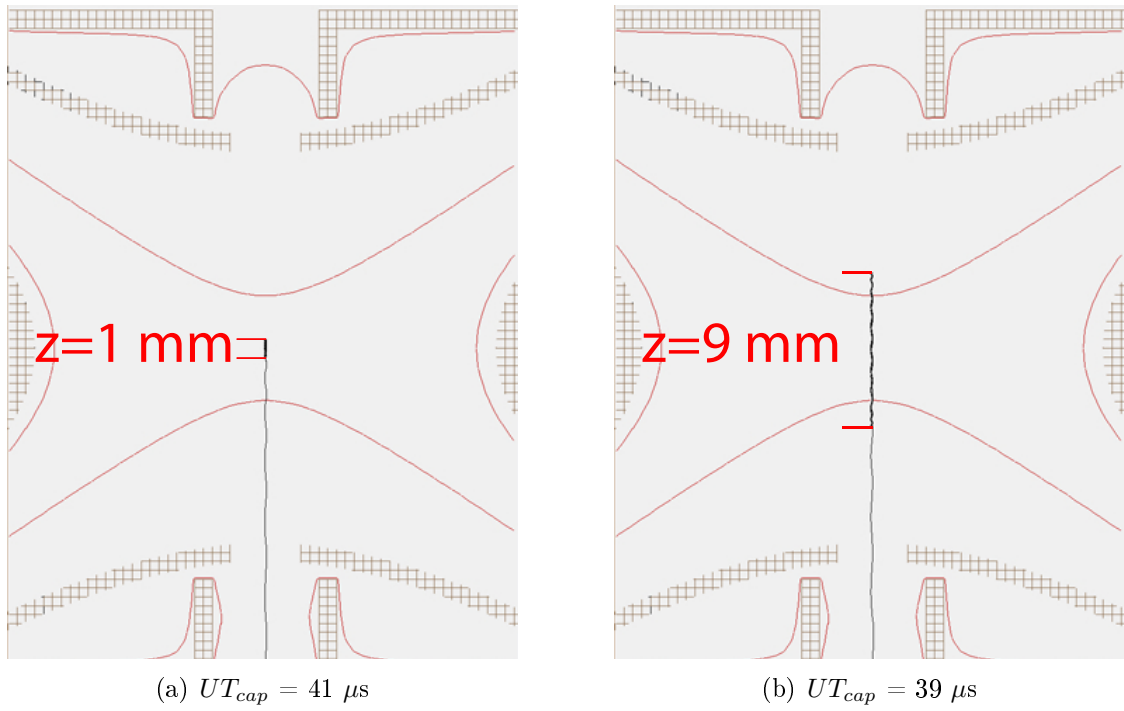
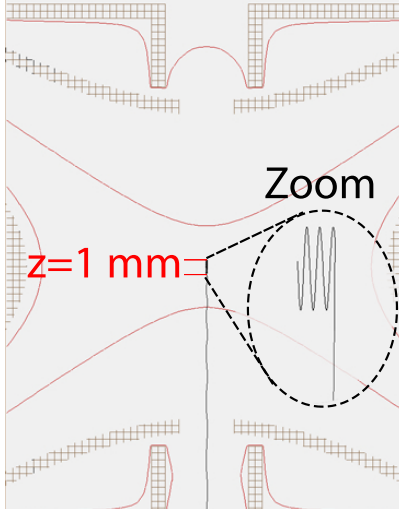
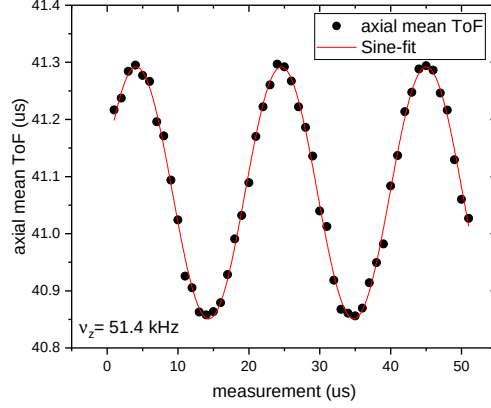


Figure A.4: Shown are the simulated ion axial deflections, resulting from different upper trap capture times UT_{cap} . (a) $z = 1 \text{ mm}$ at $UT_{cap} = 41 \mu s$ and (b) $z = 9 \text{ mm}$ at $UT_{cap} = 39 \mu s$, respectively, $43 \mu s$, as discussed in Section 4.4.1.

A.5 Upper trap axial motion



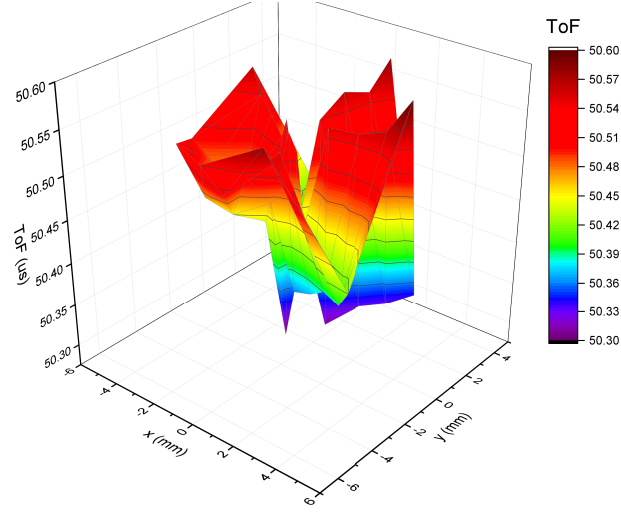
(a)



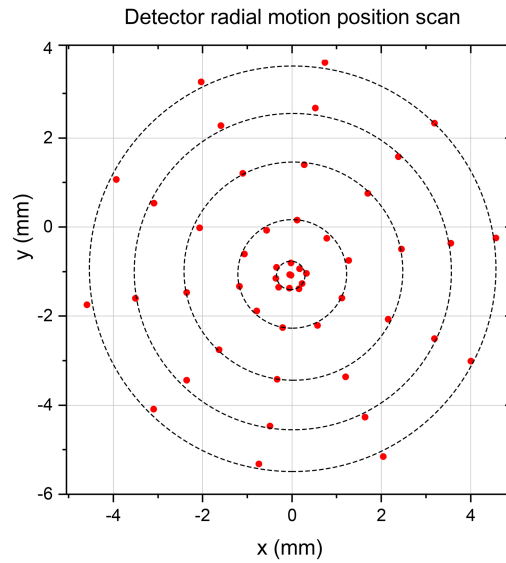
(b)

Figure A.5: The axial ion motion can be probed by use of the ToF information, recorded by the position-sensitive detector and the correct radial ion motion scan range in the order of the axial motion frequency, as presented here with a *SIMION* simulation. The ToF was recorded with an axial motion amplitude of $z = 1$ mm and a radial phase scan step width of $t_{step} = 1$ μ s. Each axial ToF position is the axial mean ToF, which is determined with 100 ions per step. The sine-fit of the axial frequency results to $\nu_z = 51.4$ kHz, which is the expected axial frequency for a mass $A = 85$ with charge $q = 1$ at ISOLTRAP, as shown in Section 2.2.3 Table 2.1. Figure (a) represents the axial motion within the trap, while (b) shows the recorded ion events on the position-sensitive detector of the projected axial motion from the trap. The recorded real ion data gives same evidence and will be published elsewhere.

A.6 Mean ToF and the unlocked magnetron motion phase scan



(a)



(b)

Figure A.6: The axial motion affects the mean ion time-of-flight, as shown in the 2D+ToF information plot in Fig. (a), while different magnetron phase positions are scanned, as highlighted with the 2D planar projection of Fig. (a) in Fig. (b). In Appendix A.5 it was demonstrated that the axial motion can be scanned and makes itself felt in an oscillation of the ion's mean ToF. Thus, the ion's motion has to be phase-locked in steps of the axial motions frequency, when the ToF information is used to define, for example, the detector/trap inclination, as presented in Section 4.4.6.

A.7 Comparison between the resolving power of ToF-ICR and PI-ICR in case of ^{129}Cd

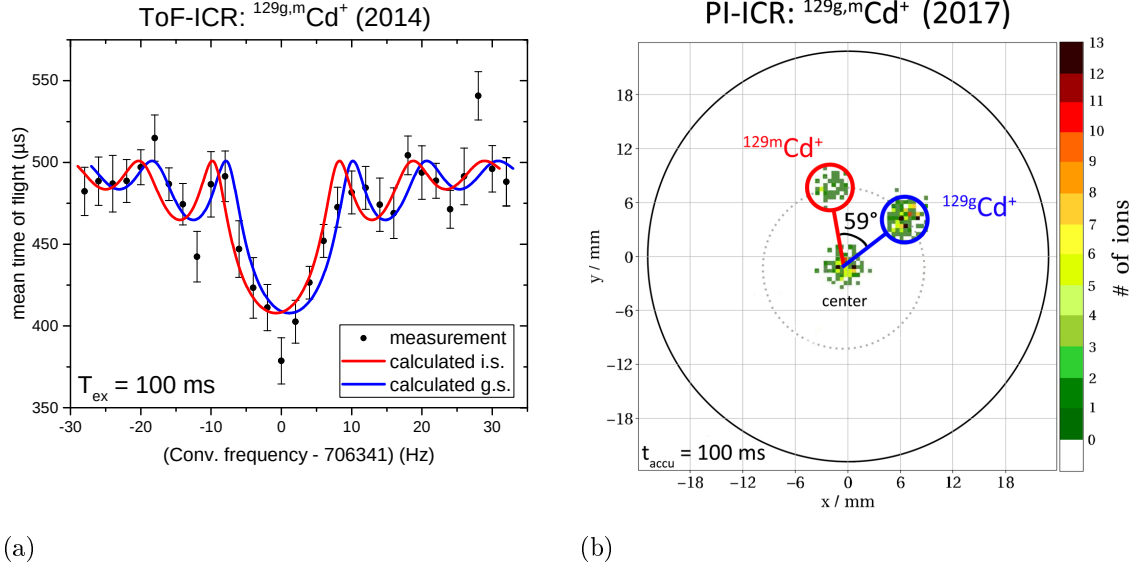


Figure A.7: When measuring $^{129}\text{Cd}^+$ in 2014 ([[AW15](#)]), as shown in (a), it becomes clear that the ToF-ICR technique cannot resolve the isomeric-state (red) and the ground-state (blue) with an excitation time of $T_{\text{ex}} = 100 \text{ ms}$. For the calculated fits in (a) an excitation energy of 350 keV has been considered. The limiting factors are the half-lives of $T_{1/2} = 151.5$ (5.7) ms for the ground-state and $T_{1/2} = 147$ (3) ms for the isomeric-state [[Aud17](#)], as well as the overall available ion yield at the time. Under the same conditions, we see the opposite in (b), where the PI-ICR technique is capable of resolving both states with an accumulation time of $t_{\text{accu}} = 100 \text{ ms}$. This accentuates the resolution of the novel PI-ICR technique compared to ToF-ICR. The results of the $^{129g,m}\text{Cd}^+$ measurement performed in 2017 will be published elsewhere.

Glossary

α	α -decay particle
$\beta^\pm$	β -decay particle
α_{ToF}	A calibration constant for the MR-ToF time of flight.
β_{ToF}	A calibration constant for the MR-ToF time of flight.
γ	emission of a photon
$\gamma_\pm$	damping constant of the radial motion
m	mass
q	charge
A	atomic number
Z	proton number
N	neutron number
E_{bin}	binding energy of a nucleon
PI-ICR	Phase-Imaging Ion-Cyclotron-Resonance technique
ToF-ICR	Time-of-Flight Ion-Cyclotron-Resonance technique
Q -Value	energy difference between mother and daughter nucleus
EC	electron capture
n_0	number of mother nuclides
$T_{1/2}$	half-life
e^-	electron
e^+	positron
A_ZX_N	mother nucleus
${}^A_ZX_N^*$	excited mother nucleus
A_ZY_N	daughter nucleus
ν_e	electron neutrino
$\bar{\nu}_e$	electron anti-neutrino
u	up-quark
d	down-quark
$W^\pm$	$W^\pm$ gauge-boson
IT	internal transition
ΔE	excess energy
m_e	mass of an electron
K, L, M	electron shell number
n	orbital shell number, principle q.n.
l	subshell orbitals, azimuthal q.n.

m_l	angular momentum, magnetic q.n.
m_s	spin projection q.n.
m_j	angular momenta correlation
q.n.	quantum number
$R_{n,l}(r)$	radial wave-function
$Y_l^m(\Theta, \Phi)$	orientation wave-function
j	total angular momentum
$m_{at}(Z, N)$	mass of an atom
$m_{nuc}(Z, N)$	mass of a nucleus
$E_{el}(Z)$	electrons binding energy
$\delta m(Z, N)$	mass defect
E_F^Z	Fermi energy of protons
E_F^N	Fermi energy of neutrons
c_0	speed of light in vacuum
E_{nuc}	binding energy of a nucleus
E_{ex}	excitation energy
eV	electron volt
$\alpha_{e.m.}$	electromagnetic fine structure constant
C.L.	confidence level
QED	quantum electrodynamics
m_p	mass of a proton
$m_{\bar{p}}$	mass of an anti-proton
h	Planck constant
N_A	Avogadro constant
GSI	GSI Helmholtz center for heavy ion research near Darmstadt
ϵ_{α_i}	single particle energy
H_{MF}	hamiltonian of the mean-field
r-process	rapid neutron capture process
$v_{LS}(r)$	spin-orbit interaction term
$v_{WS}(r)$	Wood-Saxon potential
$v_C(r)$	Coulomb potential
s	linear displacement
ϵ_0	permittivity of free space
a	surface diffuseness of a nuclide
V_0	Fermi-energy potential
$r_{0,at}$	atomic radius
Δ_n	empirical pairing-gap
S_n	one-neutron separation energy
S_{2n}	two-neutron separation energy
Π_{N_0}	$\Pi_{N_0} = (-1)^{N_0}$
UC _x	uranium carbide
E_{therm}	thermal ion energy
U_{DC}	direct current voltage
U_{rf}	radio frequency voltage

RFQ	radio-frequency quadrupole
$\mathbf{F}_\delta$	damping force
$\mathbf{v}$	velocity
δ	damping parameter
K_0	ion mobility
T_{ex}	excitation energy
p	pressure
p_N	normal pressure
T_N	normal temperature
U_{HV}	high voltage
MR-ToF MS	Multi-Reflection Time-of-Flight Mass Separator/Spectrometer
BNG	Bradbury-Nielson beam Gate
m_{IOI}	mass of the Ion-of-Interest
C_{ToF}	experimental time-of-flight ratio
m_{ref}	reference mass
$\mathfrak{R}$	mass resolving power
Δt	time-of-flight spread
Δt_{th}	thermal time spread
T_r	revolution time spread
n_r	MR-ToF revolution number
n_+	reduced cyclotron revolution number
n_-	magnetron revolution number
$\Delta\delta_E$	relative kinetic energy spread
Δt_E	ion bunch kinetic energy distribution
$\mathbf{B}$	magnetic field strength
d	characteristic trap dimension
r_0	radial trap dimension
z_0	axial direction
$\mathbf{F}_{lor}$	Lorentz force
$\mathbf{F}_{el}$	electrostatic force
ω_c, ν_c	free cyclotron frequency
ω_+, ν_+	reduced cyclotron frequency
ω_-, ν_-	magnetron frequency
ω_z, ν_z	axial frequency
U_{trap}	UT ring potential voltage
UT	upper trap
LT	lower trap
$\Phi(r,z)$	trap potential
$\bar{E}_{kin}$	particles time-averaged kinetic energy
$\bar{E}_{pot}$	particles time-averaged potential energy
ρ_+	reduced cyclotron radius
ρ_-	magnetron radius
E_+	reduced cyclotron ion energy
E_-	magnetron ion energy

E_z	axial ion energy
C_4, C_6	electrostatic field perturbation
ϵ	ellipticity
B_2	hexapole component of the magnetic field
U_{dip}	dipole excitation voltage amplitude
U_{quad}	quadrupole excitation voltage amplitude
E_{rad}	kinetic radial energy
ω_R	Rabi-frequency
T_{conv}	conversion time
FT-ICR	Fourier-Transform Ion-Cyclotron-Resonance technique
μ	magnetic moment
MCP	Multi-Channel-Plate
M_{NOI}	mass of the nuclide of interest
IOI	Ion-of-Interest
<i>ref</i>	reference
<i>u</i>	standard mass unit
ME_{exp}	experimental mass excess
ME_{lit}	literature mass excess
B.r.	Birge ratio
SM	shell model
<i>f</i>	final
t_{accu}	accumulation time
P1	magnetron motion timing pattern
P2	reduced cyclotron motion timing pattern
k_B	Boltzman constant
Φ_+	reduced cyclotron phase
Φ_-	magnetron phase
$\delta\nu_{tilt}$	frequency shift due to detector tilt
N_{ion}	amount of detected ion events
U_{DL}	delay-line detector voltages
t_{axial}	axial mean time-of-flight ion distribution
CFD	constant fraction discriminator
$\Delta\Phi$	phase difference
r_{det}	ion radius projected onto the detector
$\bar{\Phi}$	average phase
$\delta\Phi_{slow}$	phase shift caused by slow voltage fluctuation
$\delta\nu_{c,U}$	cyclotron frequency uncertainty due to voltage fluctuation
$\delta\nu_{c,B}$	cyclotron frequency uncertainty due to magnetic field fluctuation
δ_{tilt}	detector tilt
Θ	tilt of the magnetic field symmetry axis related to the electric field symmetry axis
ρ_{trap}	ion motion radius
z_{det}	distance from trap center to detector
ϕ_{trap}	angle of the ion motion in the trap

ϕ_{det}	angle of the ion motion projected onto the detector
P_B	magnetic field magnification factor
U_{drift}	drift voltage
C_{real}	real center of the projected ions
$ \alpha_{\pm}(t) $	complex amplitudes
V_{pp}	peak-to-peak voltage
δ_{dt}	detuned frequency
Δt_{delay}	delayed time difference
FIFO	fast in/fast out module
TDR	time domain reflectometer
FDR	frequency domain reflectometer
FPGA	Fast Programmable Gate Array
TDC	time-to-digital converter
TTL	transistor-transistor logic
DLD	delay-line detector

Bibliography

- [ABG48] R. A. Alpher, H. Bethe, and G. Gamow. “The Origin of Chemical Elements,” in: *Physical Review* 73 (Apr. 1948), pp. 803–804.
- [Ade16] P. A. R. Ade et al. “Planck 2015 results. XIII. Cosmological parameters,” in: *Astron. Astrophys.* 594 (2016), A13.
- [Ago16] M. Agostini et al. “Search of Neutrinoless Double Beta Decay with the GERDA Experiment,” in: *Nuclear and Particle Physics Proceedings* 273–275 (2016), pp. 1876 –1882.
- [Ago17] M. Agostini et al. “Background-free search for neutrinoless double- β decay of ^{76}Ge with GERDA,” in: *Nature* 544.7648 (2017), p. 47.
- [AGT07] M. Arnould, S. Goriely, and K. Takahashi. “The r-process of stellar nucleosynthesis: Astrophysics and nuclear physics achievements and mysteries,” in: *Physics Reports* 450.4–6 (2007), pp. 97 –213.
- [Ahm02] Q. R. Ahmad et al. “Direct Evidence for Neutrino Flavor Transformation from Neutral-Current Interactions in the Sudbury Neutrino Observatory,” in: *Phys. Rev. Lett.* 89 (1 2002), p. 011301.
- [AK64] P. W. Anderson and Y. B. Kim. “Hard Superconductivity: Theory of the Motion of Abrikosov Flux Lines,” in: *Rev. Mod. Phys.* 36 (1 1964), pp. 39–43.
- [Alp15] B. Alpert et al. “HOLMES,” in: *Eur. Phys. J. C* 75 (2015), p. 112.
- [And00] A. N. Andreyev et al. “A triplet of differently shaped spin-zero states in the atomic nucleus ^{186}Pb ,” in: *Nature* 405.6785 (2000), p. 430.
- [And11] B. Andreas et al. “Determination of the Avogadro Constant by Counting the Atoms in a ^{28}Si Crystal,” in: *Phys. Rev. Lett.* 106 (3 2011), p. 030801.
- [And62] P. W. Anderson. “Theory of Flux Creep in Hard Superconductors,” in: *Phys. Rev. Lett.* 9 (7 1962), pp. 309–311.
- [And82] T. Andersen et al. In: *Phys. Lett. B* 113 (1982), p. 72.
- [Asc16] P. Ascher et al. “ISOLTRAP’s double in trap decay studies,” in: *In preparation* (2016).
- [Ast20] F.W. Aston. “LIX. The mass-spectra of chemical elements,” in: *Philosophical Magazine Series 6* 39.233 (1920), pp. 611–625.

- [Ata16] D. Atanasov. “Precision mass measurements for studies of nucleosynthesis via the rapid neutron-capture process,” in: *PhD Thesis, Ruprecht-Karls-Universität, Heidelberg* (2016).
- [Aud17] G. Audi et al. “The NUBASE2016 evaluation of nuclear properties,” in: *Chinese physics C* 41.3 (2017), p. 030001.
- [AW15] D. Atanasov, A. Welker, et al. “Precision Mass Measurements of $^{129-131}\text{Cd}$ and Their Impact on Stellar Nucleosynthesis via the Rapid Neutron Capture Process,” in: *Phys. Rev. Lett.* 115 (23 2015), p. 232501.
- [AW17] N.A. Althubiti, A. Welker, et al. “Spectroscopy of the long-lived excited state in the neutron-deficient nuclides Po 195, 197, 199 by precision mass measurements,” in: *Physical Review C* 96.4 (2017), p. 044325.
- [BB36] H. A. Bethe and R. F. Bacher. “Nuclear Physics A. Stationary States of Nuclei,” in: *Rev. Mod. Phys.* 8 (2 1936), pp. 82–229.
- [BDN13] K. Blaum, J. Dilling, and W. Nörtershäuser. “Precision atomic physics techniques for nuclear physics with radioactive beams,” in: *Physica Scripta* 2013.T152 (2013), p. 014017.
- [Bec09] D. Beck et al. “Electric and magnetic field optimization procedure for Penning trap mass spectrometers,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 598.2 (2009), pp. 635–641.
- [Bec96] A. H. Becquerel. “Sur les radiations émises par phosphorescence,” in: *Comptes rendus* 122 (1896), pp. 420–421.
- [Ben00] M. Bender et al. “Pairing gaps from nuclear mean-field models,” in: *The European Physical Journal A* 8.1 (2000), pp. 59–75.
- [Ben08] M. Bender et al. “Collectivity-induced quenching of signatures for shell closures,” in: *Phys. Rev. C* 78 (5 2008), p. 054312.
- [BG86] L. S. Brown and G. Gabrielse. “Geonium theory: Physics of a single electron or ion in a Penning trap,” in: *Rev. Mod. Phys.* 58 (1 1986), pp. 233–311.
- [BHR03] M. Bender, P.-H. Heenen, and P.-G. Reinhard. “Self-consistent mean-field models for nuclear structure,” in: *Rev. Mod. Phys.* 75 (1 2003), pp. 121–180.
- [Bir32] R. T. Birge. “The Calculation of Errors by the Method of Least Squares,” in: *Phys. Rev.* 40 (2 1932), pp. 207–227.
- [Bla03] K. Blaum et al. “Recent developments at ISOLTRAP: towards a relative mass accuracy of exotic nuclei below 10^{-8} ,” in: *Journal of Physics B: Atomic, Molecular and Optical Physics* 36.5 (2003), p. 921.
- [Bla06] K. Blaum. “High-accuracy mass spectrometry with stored ions,” in: *Phys. Rep.* 425 (2006), p. 1.

- [Bla08] A. Blanc. “Recherches sur les mobilités des ions dans les gaz,” in: *Journal de Physique Théorique et Appliquée* 7.1 (1908), pp. 825–839.
- [BN36] N. E. Bradbury and R. A. Nielsen. “Absolute Values of the Electron Mobility in Hydrogen,” in: *Phys. Rev.* 49 (5 1936), pp. 388–393.
- [BNW10] K. Blaum, Yu. N. Novikov, and G. Werth. “Penning traps as a versatile tool for precise experiments in fundamental physics,” in: *Contemporary Physics* 51.2 (2010), pp. 149–175.
- [Boe14] Ch. Boehm et al. In: *Phys. Rev. C* 90 (4 2014), p. 044307.
- [Bog14] S. K. Bogner et al. “Nonperturbative Shell-Model Interactions from the In-Medium Similarity Renormalization Group,” in: *Phys. Rev. Lett.* 113 (14 2014), p. 142501.
- [Bol01] G. Bollen. “Mass measurements of short-lived nuclides with ion traps,” in: *Nuclear Physics A* 693.1-2 (2001), pp. 3–18.
- [Bol90] G. Bollen et al. “The accuracy of heavy-ion mass measurements using time of flight-ion cyclotron resonance in a Penning trap,” in: *Journal of Applied Physics* 68.9 (1990), pp. 4355–4374.
- [Bol94] G. Bollen. “Präzisionsbestimmung atomarer Massen,” in: *Habilitationsschrift* (1994).
- [Bor16] M.J.G. Borge. “Highlights of the ISOLDE facility and the HIE-ISOLDE project,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 376 (2016), pp. 408–412.
- [Bud67] G. I. Budker. “An effective method of damping particle oscillations in proton and antiproton storage rings,” in: *Soviet Atomic Energy* 22.5 (1967), pp. 438–440.
- [Cau05] E. Caurier et al. “The shell model as a unified view of nuclear structure,” in: *Rev. Mod. Phys.* 77 (2 2005), pp. 427–488.
- [Cha32] J. Chadwick. “The Existence of a Neutron,” in: *Proc. Roy. Soc.* 16.830 (1932), pp. 692–708.
- [col16] The ISOLDE collaboration. “ISOLDE yield database,” in: *ISOLDE yield database* (2016).
- [Coo14] R. Cooke et al. “Precision measures of the primordial abundance of deuterium,” in: *Astrophys. J.* 781.1 (2014), p. 31.
- [CQW15] S.A. Changizi, C. Qi, and R. Wyss. “Empirical pairing gaps, shell effects, and di-neutron spatial correlation in neutron-rich nuclei,” in: *Nuclear Physics A* 940 (2015), pp. 210–226.
- [Cro14] M.P. Croce et al. In: *Low Temp. Phys.* 176 (2014), p. 1009.
- [Cyb16] R. H. Cyburt et al. “Dependence of X-Ray Burst Models on Nuclear Reaction Rates,” in: *The Astrophysical Journal* 830.2 (2016), p. 55.

- [Dah00] D. A. Dahl. “simion for the personal computer in reflection,” in: *International Journal of Mass Spectrometry* 200.1 (2000). Volume 200: The state of the field as we move into a new millenium, pp. 3–25.
- [Deh90] H. Dehmelt. “Experiments with an isolated subatomic particle at rest,” in: *Rev. Mod. Phys.* 62 (3 1990), pp. 525–530.
- [Dem18] A. J. Dempster. “A new Method of Positive Ray Analysis,” in: *Phys. Rev.* 11 (4 1918), pp. 316–325.
- [Die11] C. Diehl et al. “Progress with the MPIK/UW-PTMS in Heidelberg,” in: *Hyperfine Interactions* 199.1 (2011), pp. 291–300.
- [DiS13] J. DiSciaccia et al. “One-Particle Measurement of the Antiproton Magnetic Moment,” in: *Phys. Rev. Lett.* 110 (13 2013), p. 130801.
- [DRL16] A. De Rújula and M. Lusignoli. “The calorimetric spectrum of the electron-capture decay of ^{163}Ho . The spectral endpoint region,” in: *J. of High. E. Phys.* (2016), p. 15.
- [Duf95] J. Duffo et al. “Microscopic mass formulas,” in: *Phys. Rev. C* 52 (1 1995), R23–R27.
- [Dyc99] R. Van Dyck et al. “Ultrastable superconducting magnet system for a Penning trap mass spectrometer,” in: *Review of scientific instruments* 70.3 (1999), pp. 1665–1671.
- [EAM88] E. W. McDaniel E. A. Mason. *Transport Properties of Ions in Gases*. 99th ed. Wiley-Interscience, 1988.
- [Eit09] G. Eitel et al. “Position-sensitive ion detection in precision Penning trap mass spectrometry,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 606.3 (2009), pp. 475–483.
- [Eli10] S. Eliseev et al. “Direct mass measurements of ^{194}Hg and ^{194}Au : A new route to the neutrino mass determination?,” in: *Physics Letters B* 693.4 (2010), pp. 426–429.
- [Eli13] S. Eliseev et al. “Phase-Imaging Ion-Cyclotron-Resonance Measurements for Short-Lived Nuclides,” in: *Phys. Rev. L* 110 (2013), p. 082501.
- [Eli14] S. Eliseev et al. “A phase-imaging technique for cyclotron-frequency measurements,” in: *Applied Physics B* 114.1 (2014), pp. 107–128.
- [Eli15] S. Eliseev et al. “Direct Measurement of the Mass Difference of ^{163}Ho and ^{163}Dy Solves the Q -Value Puzzle for the Neutrino Mass Determination,” in: *Phys. Rev. Lett.* 115 (6 2015), p. 062501.
- [Ell78] H. W. Ellis et al. “Transport properties of gaseous ions over a wide energy range. Part II,” in: *Atomic Data and Nuclear Data Tables* 22.3 (1978), pp. 179–217.

- [Ell84] H. W. Ellis et al. “Transport properties of gaseous ions over a wide energy range. Part III,” in: *Atomic Data and Nuclear Data Tables* 31.1 (1984), pp. 113–151.
- [Erl12] J. Erler et al. “The limits of the nuclear landscape,” in: *Nature* 486 (2012), pp. 409–512.
- [Ero13] T. Eronen et al. “High-precision atomic mass measurements for a CKM unitarity test,” in: *International Journal of Mass Spectrometry* 349–350 (2013), pp. 69–73.
- [F.A20] F. Aston. “Isotopes and Atomic Weights,” in: *Nature* 105 (1920), pp. 617–619.
- [Fae15] A. Faessler et al. “The calorimetric spectrum of the electron-capture decay of ^{163}Ho . The spectral endpoint region,” in: *Phys. Rev. C* 92 (2015), p. 045505.
- [Fea35] G. Fea. “Tabelle riassuntive e bibliografia delle trasmutazioni artificiali,” in: *Il Nuovo Cimento (1924-1942)* 12.6 (1935), pp. 368–406.
- [Fed12] V. N. Fedosseev et al. “Upgrade of the resonance ionization laser ion source at ISOLDE on-line isotope separation facility: New lasers and new ion beams,” in: *Rev. Sci. Instrum.* 83.2 (2012), p. 020000.
- [Fed95] V. N. Fedoseyev et al. “Study of short-lived silver isotopes with a laser ion source,” in: *Zeitschrift für Physik A Hadrons and Nuclei* 353.1 (1995), pp. 9–10.
- [Fix09] D. J. Fixsen. “The Temperature of the Cosmic Microwave Background,” in: 707 (Dec. 2009), pp. 916–920.
- [Fla09] K. T. Flanagan et al. “Nuclear Spins and Magnetic Moments of $^{71,73,75}\text{Cu}$: Inversion of $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ Levels in ^{75}Cu ,” in: 103 (14 2009), p. 142501.
- [FT15] S. W. Finch and W. Tornow. “Search for neutrinoless double-electron capture of ^{156}Dy ,” in: *Phys. Rev. C* 92 (6 2015), p. 065503.
- [Fuk98] Y. Fukuda et al. “Evidence for Oscillation of Atmospheric Neutrinos,” in: *Phys. Rev. Lett.* 81 (8 1998), p. 1562.
- [Gab09] G. Gabrielse. “The true cyclotron frequency for particles and ions in a Penning trap,” in: *International Journal of Mass Spectrometry* 279.2–3 (2009), pp. 107–112.
- [Gam30] G. Gamow. *Mass Defect Curve and Nuclear Constitution*. Vol. 126. Proceedings Series A. Royal Society of London, 1930, pp. 632–644.
- [Gas] L. Gastaldo et al. In: ().
- [Gat97] F. Gatti et al. “Calorimetric measurement of the ^{163}Ho spectrum by means of a cryogenic detector,” in: *Physics Letters B* 398.3 (1997), pp. 415–419.

- [Gau09] L. Gaudefroy et al. “Collective structure of the $N = 40$ isotones,” in: *Phys. Rev. C* 80.6 (2009), p. 064313.
- [Geo07] S. George et al. “The Ramsey method in high-precision mass spectrometry with Penning traps: Experimental results,” in: *International Journal of Mass Spectrometry* 264.2–3 (2007), pp. 110–121.
- [Geo09] S. George. “First Ramsey-type mass measurements with ISOLTRAP and design studies of the new PENTATRAP project,” in: *PhD Thesis, Johannes Gutenberg-Universität, Mainz* (2009).
- [Geo11] S. George et al. “Damping effects in Penning trap mass spectrometry,” in: *International Journal of Mass Spectrometry* 299.2–3 (2011), pp. 102–112.
- [GG74] H. Georgi and S. L. Glashow. “Unity of All Elementary-Particle Forces,” in: *Phys. Rev. Lett.* 32 (8 1974), pp. 438–441.
- [GK66] G. T. Garvey and I. Kelson. “New Nuclidic Mass Relationship,” in: *Phys. Rev. Lett.* 16 (5 1966), pp. 197–200.
- [GM62] M. Gell-Mann. “Symmetries of Baryons and Mesons,” in: *Physical Review* 125 (Feb. 1962), pp. 1067–1084.
- [Gon14] M. Goncharov. “High-precision Penning-trap mass spectrometry at SHIP-TRAP and PENTATRAP for neutrino physics research.” In: *PhD Thesis, Ruprecht-Karls-Universität, Heidelberg* (2014).
- [Gor16] S. Goriely et al. “Further explorations of Skyrme-Hartree-Fock-Bogoliubov mass formulas. XVI. Inclusion of self-energy effects in pairing,” in: *Physical Review C* 93.3 (2016), p. 034337.
- [Got14] A. Gottberg et al. “Experimental tests of an advanced proton-to-neutron converter at ISOLDE-CERN,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 336 (2014), pp. 143–148.
- [GR16] R. F. Garcia Ruiz et al. “Unexpectedly large charge radii of neutron-rich calcium isotopes,” in: *Nature* 536 (2016), pp. 594–598.
- [Gro09] C. J. Gross et al. “Decay spectroscopy of $^{75,79}\text{Cu}$, $^{71,81}\text{Zn}$ and $^{83,85}\text{Ga}$,” in: *Acta Phys. Polonica B* 40.3 (2009), pp. 447–455.
- [Gro17] R. P. de Groote et al. “Dipole and quadrupole moments of $^{73-78}\text{Cu}$ as a test of the robustness of the $Z = 28$ shell closure near ^{78}Ni ,” in: *Phys. Rev. C* 96 (4 2017), p. 041302.
- [Grä80] G. Gräff et al. “A direct determination of the proton electron mass ratio,” in: *Z. Phys. A* 297 (1980), pp. 35–39.
- [Gut81] A. H. Guth. “Inflationary universe: A possible solution to the horizon and flatness problems,” in: *Phys. Rev. D* 23 (2 1981), pp. 347–356.

- [GXM93] S. Guan, X. Xiang, and A. G. Marshall. “Axial and radial ion cloud compression: coupling of magnetron and cyclotron motion to axial motion in a segmented cubic Fourier transform ion cyclotron resonance ion trap,” in: *International Journal of Mass Spectrometry and Ion Processes* 124.1 (1993), pp. 53–67.
- [Hag16] G. Hagen et al. “Structure of ^{78}Ni from First-Principles Computations,” in: *Phys. Rev. Lett.* 117 (17 2016), p. 172501.
- [Hag92] E. Hagebo et al. “New production systems at ISOLDE,” in: *Nuc. Inst. and Meth. in Phys. Res. Sec. B* 70 (1992), p. 165.
- [Hau06] P. A. Hausladen et al. “Opportunistic mass measurements at the Holifield Radioactive Ion Beam Facility,” in: *Int. J. Mass Spectrom.* 251.2?3 (2006), pp. 119–124.
- [Hei17] F. Heiße et al. “High-Precision Measurement of the Proton’s Atomic Mass,” in: *Physical Review Letters* 119.3 (2017), p. 033001.
- [Her01] F. Herfurth et al. “A linear radiofrequency ion trap for accumulation, bunching, and emittance improvement of radioactive ion beams,” in: *Nuc. Inst. and Meth. in Phys. Res. Sec. A* 469.2 (2001), pp. 254–275.
- [Her22] D. Hermann. “Die Fragmente der Vorsokratiker: Band 2,” in: 2 (1922).
- [HG09] E. Marsden H. Geiger J. Harling. “On a Diffuse Reflection of the α -Particles,” in: *Proc. Roy. Soc* 82 (1909), pp. 495–500.
- [HG13] E. Marsden H. Geiger J. Harling. “The Laws of Deflexion of α Particles through Large Angles,” in: *Phil. Mag.* 25.148 (1913), pp. 604–623.
- [Hon09] M. Honma et al. “New effective interaction for f 5 pg 9-shell nuclei,” in: *Phys. Rev. C* 80.6 (2009), p. 064323.
- [Hon16] R. Hong et al. “High accuracy position response calibration method for a micro-channel plate ion detector,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 835 (2016), pp. 42–50.
- [Hor16] M. Hori et al. “Buffer-gas cooling of antiprotonic helium to 1.5 to 1.7 K, and antiproton-to-electron mass ratio,” in: *Science* 354.6312 (2016), pp. 610–614.
- [HT15] J. C. Hardy and I. S. Towner. “Superaligned $0^+ \rightarrow 0^+$ nuclear β decays: 2014 critical survey, with precise results for V_{ud} and CKM unitarity,” in: *Phys. Rev. C* 91 (2 2015), p. 025501.
- [Hü15] H.-J. Hübschmann. *Handbook of GC-MS: Fundamentals and Applications*. 3rd ed. Wiley-VCH, 2015.
- [Ily09] S. V. Ilyushkin et al. “ β decay of the $\pi f_{5/2}$ ground state of ^{77}Cu studied with 225 MeV and 0.2 MeV purified radioactive beams,” in: *Phys. Rev. C* 80 (5 2009), p. 054304.

- [Iso] “ISOLDE Yield Database,” in: (2017).
- [Ita95] W. M. Itano et al. “Cooling methods in ion traps,” in: *Physica Scripta* 1995.T59 (1995), p. 106.
- [Jag02] O. Jagutzki et al. “A broad-application microchannel-plate detector system for advanced particle or photon detection tasks: large area imaging, precise multi-hit timing information and high detection rate,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 477.1–3 (2002), pp. 244–249.
- [Jen49] J. Jensen et al. “Modellmäßige Deutung der ausgezeichneten Nukleonenzahlen im Kernbau.” In: *Naturwissenschaften* 36 (1949), pp. 155–156.
- [JH13] J. H. Jungmann and R. Heeren. “Detection systems for mass spectrometry imaging: a perspective on novel developments with a focus on active pixel detectors,” in: *Rapid Communications in Mass Spectrometry* 27.1 (2013), pp. 1–23.
- [Jus15] O. Just et al. “Comprehensive nucleosynthesis analysis for ejecta of compact binary mergers,” in: *Monthly Notices of the Royal Astronomical Society* 448.1 (2015), pp. 541–567.
- [Kal11] T. Kaltenbacher et al. “Resistive cooling circuits for charged particle traps using crystal resonators,” in: *Review of Scientific Instruments* 82.11 (2011), p. 114702.
- [Kar17] J. Karthein. “Precision mass measurements using the Phase-Imaging Ion-Cyclotron-Resonance detection technique,” in: *Master Thesis, Ruprecht-Karls-Universität, Heidelberg* (2017).
- [KD96] W. Ketterle and N.J. Van Druten. “Evaporative Cooling of Trapped Atoms”. In: ed. by Benjamin Bederson and Herbert Walther. Vol. 37. *Advances In Atomic, Molecular, and Optical Physics*. Academic Press, 1996, pp. 181–236.
- [Kel03] A. Kellerbauer et al. “From direct to absolute mass measurements: A study of the accuracy of ISOLTRAP,” in: *Eur. Phys. J D* 22 (2003), pp. 53–64.
- [Ken87] K. S. Kenneth. *Introductory Nuclear Physics*. 1987.
- [Ket14] J. Ketter et al. “First-order perturbative calculation of the frequency-shifts caused by static cylindrically-symmetric electric and magnetic imperfections of a Penning trap,” in: *International Journal of Mass Spectrometry* 358 (2014), pp. 1–16.
- [Klu07] H. J. Kluge et al. In: *Nuclear Physics News* 17.4 (2007), p. 36.
- [Klu13] H. J. Kluge. “Penning trap mass spectrometry of radionuclides,” in: *International Journal of Mass Spectrometry* 349 (2013), pp. 26–37.

- [Koe15] F. Koehler et al. “The electron mass from g -factor measurements on hydrogen-like carbon $12\text{ C }5+$,” in: *Journal of Physics B: Atomic, Molecular and Optical Physics* 48.14 (2015), p. 144032.
- [Kow12] M. Kowalska et al. “Trap-assisted decay spectroscopy with ISOLTRAP,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 689 (2012), pp. 102–107.
- [KR11] V. A. Kostelecký and N. Russell. “Data tables for Lorentz and CPT violation,” in: *Rev. Mod. Phys.* 83 (1 2011), pp. 11–31.
- [Kra91] K. L. Kratz et al. “Neutron-rich isotopes around the r-process “waiting-point” nuclei $^{79}_{29}\text{Cu}_{50}$ and $^{80}_{30}\text{Zn}_{50}$,” in: *Zeit. f. Phys. A* 340.4 (1991), pp. 419–420.
- [Kre08] M. Kretzschmar. “Calculating damping effects for the ion motion in a Penning trap,” in: *The European Physical Journal D* 48.3 (2008), pp. 313–319.
- [Kre12] M. Kretzschmar. “On the phase dependence of the interconversion of the motional modes in a Penning trap by quadrupolar excitation,” in: *International Journal of Mass Spectrometry* 309 (2012), pp. 30–38.
- [Kre13] S. Kreim et al. “Recent exploits of the ISOLTRAP mass spectrometer,” in: *Nucl. Instrum. Methods B* 317 (2013), pp. 492–500.
- [Kre92] M. Kretzschmar. “Excitation of particle motions in a Penning trap: description in the classical canonical formalism,” in: *Physica Scripta* 46.6 (1992), p. 555.
- [Kug00] E. Kugler. “The ISOLDE facility,” in: *Hyp. Int.* 129.1-4 (2000), pp. 23–42.
- [Kön95] M. König et al. “Quadrupole excitation of stored ion motion at the true cyclotron frequency,” in: *Int. J. Mass. Spectrom. Ion Process* 142 (1995), pp. 95–116.
- [Kös02] U. Köster. “Intense radioactive-ion beams produced with the ISOL method,” in: *Eur. Phys. J. A* 15.1-2 (2002), pp. 255–263.
- [Lar77] F.P. Larkins. “Semiempirical Auger-electron energies for elements $10 \leq Z \leq 100$,” in: *Atom. D. and Nucl. D. Tabl.* 20 (1977), p. 311.
- [Las17] D. Lascar et al. “Precision mass measurements of $^{125,127}\text{Cd}$ isotopes and isomers approaching the $N = 82$ closed shell,” in: *Phys. Rev. C* 96 (4 2017), p. 044323.
- [Law30] E. O. Lawrence. In: *The National Academy Of Sciences* 72.1867 (1930), pp. 372–378.
- [LCN13] Y. Li, X. Chen, and G. Ni. “Analysis and modeling on noise factor of microchannel plate”. In: International Society for Optics and Photonics. 2013, 90430Y–90430Y.

- [Leh16] B. Lehnert et al. “A search for the radiative neutrinoless double-electron capture of ^{58}Ni ,” in: *Journal of Physics G: Nuclear and Particle Physics* 43.6 (2016), p. 065201.
- [Lit13] Yu. A. Litvinov et al. “Nuclear physics experiments with ion storage rings,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 317, Part B (2013), pp. 603–616.
- [LL32] E. O. Lawrence and M. S. Livingston. “The Production of High Speed Light Ions Without the Use of High Voltages,” in: *Phys. Rev.* 40 (1 1932), pp. 19–35.
- [LM99] D. Lunney and R. B. Moore. “Cooling of mass-separated beams using a radiofrequency quadrupole ion guide,” in: *International Journal of Mass Spectrometry* 190–191 (1999), pp. 153–160.
- [LPT03] D. Lunney, J. M. Pearson, and C. Thibault. “Recent trends in the determination of nuclear masses,” in: *Rev. Mod. Phys.* 75 (3 2003), pp. 1021–1082.
- [Lun17] D. Lunney et al. “Extending and refining the nuclear mass surface with ISOLTRAP,” in: *Journal of Physics G: Nuclear and Particle Physics* 44.6 (2017), p. 064008.
- [Maj06] F. Major et al. *Charged particle traps: physics and techniques of charged particle field confinement*. Vol. 37. Springer Science & Business Media, 2006.
- [Mam73] B. A. Mamyrin et al. “Massreflectron: A new nonmagnetic time-of-flight high-resolution mass-spectrometer,” in: *Sov. Phy. JETP-USSR* 64 (1973), 82–89.
- [Man13] V. Manea et al. “Collective degrees of freedom of neutron-rich $A \approx 100$ nuclei and the first mass measurement of the short-lived nuclide ^{100}Rb ,” in: *Phys. Rev. C* 88 (5 2013), p. 054322.
- [Man15] V. Manea. *Binding Energy of Strongly Deformed Radionuclides: Penning-trap Mass Spectrometry and Mean-field Theoretical Studies*. Springer, 2015.
- [Mar13] B. A. Marsh et al. “New developments of the in-source spectroscopy method at RILIS/ISOLDE,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 317, Part B (2013), pp. 550–556.
- [Mar18] B. A. Marsh et al. “Mercurial nuclei get back in shape: In preparation,” in: *Nature* (2018).
- [May48] M. G. Mayer. “On Closed Shells in Nuclei,” in: *Phys. Rev.* 74 (3 1948), pp. 235–239.

- [MCB98] M., P. Curie, and G. Bémont. “Sur une nouvelle substance fortement radio-active, contenue dans la pechblende,” in: *Comptes rendus* 127 (1898), pp. 1215–1217.
- [Men69] D. Mendelejew. “Über die Beziehungen der Eigenschaften zu den Atomgewichten der Elemente,” in: *Z. für Chem.* 12 (1869), pp. 405–406.
- [MH34] J. Mattauch and R. Herzog. “Über einen neuen Massenspektrographen,” in: *Zeitschrift für Physik* 89.11 (1934), pp. 786–795.
- [MHJ98] A. G. Marshall, C. L. Hendrickson, and G. S. Jackson. “Fourier transform ion cyclotron resonance mass spectrometry: a primer,” in: *Mass spectrometry reviews* 17.1 (1998), pp. 1–35.
- [MJ55] M. G. Mayer and J. H. D. Jensen. *Elementary Theory of Nuclear Shell Structure*. Structure of matter series. John Wiley & Sons, 1955.
- [MK02] T. Mayer-Kuckuk. *Kernphysik: Eine Einführung*. 7th ed. Teubner Studienbücher Physik. Vieweg+Teubner Verlag, 2002.
- [MS66] W. D. Myers and W. J. Swiatecki. “Nuclear masses and deformations,” in: *Nuclear Physics* 81.1 (1966), pp. 1–60.
- [Mue16a] C. Mueller et al. “Alpha-PET with terbium-149: evidence and perspectives for radiotheragnostics,” in: *EJNMMI Radiopharmacy and Chemistry* 1.1 (2016), p. 5.
- [Mue16b] C. Mueller et al. “Preclinical in vivo application of (152)Tb-DOTANOC: a radiolanthanide for PET imaging,” in: *EJNMMI Res* 6 (2016). 189[PII], p. 35.
- [Muk08] M. Mukherjee et al. “ISOLTRAP: An on-line Penning trap for mass spectrometry on short-lived nuclides,” in: *The European Physical Journal A-Hadrons and Nuclei* 35.1 (2008), pp. 1–29.
- [Mye13] E. G. Myers. “The most precise atomic mass measurements in Penning traps,” in: *International Journal of Mass Spectrometry* 349–350 (2013). 100 years of Mass Spectrometry, pp. 107–122.
- [Mye15] E. G. Myers et al. “Atomic Masses of Tritium and Helium-3,” in: *Phys. Rev. Lett.* 114 (1 2015), p. 013003.
- [Nai12] S. Naimi et al. “Surveying the $N = 40$ island of inversion with new manganese masses,” in: *Phys. Rev. C* 86 (1 2012), p. 014325.
- [Nes14] D. A. Nesterenko et al. “Direct determination of the atomic mass difference of ^{187}Re and ^{187}Os for neutrino physics and cosmochemistry,” in: *Phys. Rev. C* 90 (4 2014), p. 042501.
- [Nie05] H. B. Niemann et al. “The abundances of constituents of Titan’s atmosphere from the GCMS instrument on the Huygens probe,” in: *Nature* 438.7069 (2005), pp. 779–784.

- [NL18] Z. M. Niu and H. Z. Liang. “Nuclear mass predictions based on Bayesian neural network approach with pairing and shell effects,” in: *Physics Letters B* 778 (2018), pp. 48–53.
- [Now16] F. Nowacki et al. “Shape Coexistence in ^{78}Ni as the Portal to the Fifth Island of Inversion,” in: *Phys. Rev. Lett.* 117 (1 2016), p. 272501.
- [OJM07] R. Oechslin, H.-T. Janka, and A. Marek. “Relativistic neutron star merger simulations with non-zero temperature equations of state-I. Variation of binary parameters and equation of state,” in: *Astronomy & Astrophysics* 467.2 (2007), pp. 395–409.
- [Oli17] L. Olivier et al. “Persistence of the $Z=28$ Shell Gap Around $\text{Ni } 78$: First Spectroscopy of $\text{Cu } 79$,” in: *Physical review letters* 119.19 (2017), p. 192501.
- [OOC11] H.M. O’Keeffe, E. O’Sullivan, and M.C. Chen. “Scintillation decay time and pulse shape discrimination in oxygenated and deoxygenated solutions of linear alkylbenzene for the SNO+ experiment,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 640.1 (2011), pp. 119–122.
- [Orl15] R. Orlandi et al. “Single-neutron orbits near ^{78}Ni : Spectroscopy of the $N=49$ isotope ^{79}Zn ,” in: *Phys. Lett. B* 740 (2015), pp. 298–302.
- [Ots05] T. Otsuka et al. “Evolution of Nuclear Shells due to the Tensor Force,” in: *Phys. Rev. Lett.* 95 (23 2005), p. 232502.
- [Ots06] T. Otsuka et al. “Mean Field with Tensor Force and Shell Structure of Exotic Nuclei,” in: *Phys. Rev. Lett.* 97 (16 2006), p. 162501.
- [OW08] E. W. Otten and Ch. Weinheimer. “Neutrino mass limit from tritium β decay,” in: *Reports on Progress in Physics* 71.8 (2008), p. 086201.
- [Par16] H.-I. Park. “Precise test of the unitarity of the CKM matrix via superallowed nuclear beta decay”. In: *APS April Meeting Abstracts*. Mar. 2016.
- [Pat09] N. Patronis et al. “ β -decay study of ^{77}Cu ,” in: *Phys. Rev. C* 80 (3 2009), p. 034307.
- [Pau25] W. Pauli. “Über den Zusammenhang des Abschlusses der Elektronengruppen im Atom mit der Komplexstruktur der Spektren,” in: *Zeitschrift für Physik* 31.1 (1925), pp. 765–783.
- [Pau90] W. Paul. “Electromagnetic traps for charged and neutral particles,” in: *Rev. Mod. Phys.* 62 (3 1990), pp. 531–540.
- [Pen36] F.M. Penning. “Verzögerungen bei der zündung von gasgefüllten photozellen im dunkeln,” in: *Physica* 3.6 (1936), pp. 563–568.

- [Pet86] Z. L. Petrovic. “The application of Blanc’s law to the determination of the diffusion coefficients for thermal electrons in gases,” in: *Australian journal of physics* 39.2 (1986), pp. 237–248.
- [Pf2] M. Pfützner et al. “Radioactive decays at limits of nuclear stability,” in: *Rev. Mod. Phys.* 84 (2 2012), pp. 567–619.
- [Pia17] E. Pian et al. “Spectroscopic identification of r-process,” in: *Nature* 551 (2017), pp. 67–70.
- [Pie49] J. R. Pierce. “Theory and Design of Electron Beams,” in: (1949).
- [Por01] J. V. Porto. “Series solution for the image charge fields in arbitrary cylindrically symmetric Penning traps,” in: *Phys. Rev. A* 64 (2 2001), p. 023403.
- [Por15] T. Porobić et al. “Space-charge effects in Penning ion traps,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 785 (2015), pp. 153–162.
- [Pov17] B. Povh et al. “Cosmology-The Early Universe”. In: *Scattering and Structures*. Springer, 2017, pp. 207–218.
- [PS53] W. Paul and H. Steinwedel. “Notizen: Ein neues Massenspektrometer ohne Magnetfeld,” in: *Zeitschrift Naturforschung Teil A* 8 (July 1953), pp. 448–450.
- [PSB15] F. Priester, M. Sturm, and B. Bornschein. “Commissioning and detailed results of KATRIN inner loop tritium processing system at Tritium Laboratory Karlsruhe,” in: *Vacuum* 116 (2015), pp. 42–47.
- [Pud97] B. S. Pudliner et al. “Quantum Monte Carlo calculations of nuclei with *Alt*; 7,” in: *Phys. Rev. C* 56 (4 1997), pp. 1720–1750.
- [Ran12] P.C.-O. Ranitzsch et al. In: *J. Low Temp. Phys.* 167 (2012), p. 1004.
- [Red13] M. Redshaw et al. “LEBIT II: Upgrades and developments for high precision Penning trap mass measurements with rare isotopes,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 317 (2013), pp. 510–516.
- [Rep12] J. Repp et al. “PENTATRAP: a novel cryogenic multi-Penning-trap experiment for high-precision mass measurements on highly charged ions,” in: *Applied Physics B* 107.4 (2012), pp. 983–996.
- [RH97] H. Raimbault-Hartmann et al. In: *Nucl. Instr. Meth. B* 126.1–4 (1997), pp. 378–382.
- [Rin07] R. Ringle et al. “Octupolar excitation of ion motion in a Penning trap—A study performed at {LEBIT},” in: *International Journal of Mass Spectrometry* 262.1–2 (2007), pp. 33–44.
- [Roe17a] Roentdek. “RoentDek amplifier manual for FAMP3, FAMP6 and FAMP8,” in: (2017).

- [Roe17b] Roentdek. “RoentDek DLD/HEX delay-line detector manual.” In: (2017).
- [Ros15] M. Rosenbusch et al. “Probing the $N = 32$ Shell Closure below the Magic Proton Number $Z = 20$: Mass Measurements of the Exotic Isotopes $^{52,53}\text{K}$,” in: *Phys. Rev. Lett.* 114 (20 2015), p. 202501.
- [Rot12] A. Roth. *Vacuum technology*. Elsevier, 2012.
- [Rot16] S. Rothe et al. “Laser ion beam production at CERN-ISOLDE: New features – More possibilities,” in: *Nuc. Inst. and Meth. in Phys. Res. Sec. B* 376 (2016), p. 91.
- [Rou12] C. Roux et al. “The trap design of PENTATRAP,” in: *Applied Physics B* 107.4 (2012), pp. 997–1005.
- [RSB13] R. Ringle, S. Schwarz, and G. Bollen. “Penning trap mass spectrometry of rare isotopes produced via projectile fragmentation at the LEBIT facility,” in: *International Journal of Mass Spectrometry* 349 (2013), pp. 87–93.
- [Rut03] E. Rutherford. “XV. The magnetic and electric deviation of the easily absorbed rays from radium,” in: *Phil. Mag. Series 6* 5.26 (1903), pp. 177–187.
- [Rut11] E. Rutherford. “The Scattering of α and β Particles by Matter and the Structure of the Atom,” in: *Phil. Mag. Series 6* 21 (1911), pp. 669–688.
- [Rut20] E. Rutherford. “Bakerian Lecture. Nuclear Constitution of Atoms,” in: *Proceedings of the Royal Society of London Series A* 97 (July 1920), pp. 374–400.
- [Rut99] E. Rutherford. “VIII. Uranium radiation and the electrical conduction produced by it,” in: *Philosophical Magazine Series 5* 47.284 (1899), pp. 109–163.
- [Rúj82] A. De Rújula et al. “Calorimetric measurements of ^{163}Ho decay as tools to determine the electron neutrino mass,” in: *Phys. Lett.* 118 (1982), p. 429.
- [Sah17] E. Sahin et al. “Shell Evolution towards ^{78}Ni : Low-Lying States in ^{77}Cu ,” in: *Phys. Rev. Lett.* 118 (24 2017), p. 242502.
- [Sat97] W. Satuła et al. “On the origin of the Wigner energy,” in: *Physics Letters B* 407.2 (1997), pp. 103–109.
- [Sav91] G. Savard et al. “A new cooling technique for heavy ions in a Penning trap,” in: *Phys. Lett. A* 158.5 (1991), pp. 247–252.
- [SBB87] P. T. Springer, C. L. Bennett, and P. A. Baisden. “Measurement of the neutrino mass using the inner bremsstrahlung emitted in the electron-capture decay of ^{163}Ho ,” in: *Phys. Rev. A* 35 (2 1987), pp. 679–689.
- [Sch05] H. Schatz et al. “The half-life of the doubly-magic r-process nucleus ^{78}Ni ,” in: *The European Physical Journal A - Hadrons and Nuclei* 25.1 (2005), pp. 639–642.

- [Sch17] M. Schioppo et al. “Ultrastable optical clock with two cold-atom ensembles,” in: *Nat Photon* 11.1 (2017). Letter, pp. 48–52.
- [SO17] H. Schatz and W. J. Ong. “Dependence of X-ray Burst Models on Nuclear Masses,” in: *The Astrophysical Journal* 844 (2 2017).
- [Sok67] A. Sokolov et al. “Induced and Spontaneous Emission in Crossed Fields,” in: *Optics and Spectroscopy* 22 (Jan. 1967), p. 1.
- [Sor08] O. Sorlin et al. “Nuclear magic numbers: New features far from stability,” in: *Progr. Part. Nucl. Phys.* 61 (2008), p. 602.
- [Sta13] J. Stanja et al. “Mass spectrometry and decay spectroscopy of isomers across the $Z = 82$ shell closure,” in: *Phys. Rev. C* 88 (5 2013), p. 054304.
- [Ste13] D. Steppenbeck et al. “Evidence for a new nuclear ‘magic number’ from the level structure of ^{54}Ca ,” in: *Nature* 502 (2013), pp. 207–210.
- [Ste16] D. Steppenbeck et al. “Low-lying Structures Of Exotic Sc Isotopes And The Evolution Of The $N = 34$ Subshell Closure”. In: 2016.
- [Ste46] W. E. Stephens. “A pulsed mass spectrometer with time dispersion,” in: *Phys. Rev.* 69 (1946), p. 691.
- [Sto11] M. Stock. “The watt balance: determination of the Planck constant and redefinition of the kilogram,” in: *Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences* 369.1953 (2011), pp. 3936–3953.
- [Suh07] J. Suhonen. *From Nucleons to Nucleus: Concepts of Microscopic Nuclear Theory*. 1st ed. Theoretical and Mathematical Physics. Springer-Verlag Berlin Heidelberg, 2007.
- [Tho13] J. J. Thomson. “Rays of positive electricity, and their application to chemical analyses,” in: *Monographs on Physics* 1 (1913).
- [Tho97] J. J. Thomson. “XL. Cathode Rays,” in: *Philosophical Magazine Series* 5 44.269 (1897), pp. 293–316.
- [Tre60] Werner Tretner. “An electrostatic mass spectroscope,” in: *Vacuum* 10.1 (1960), pp. 31–34.
- [Ulm13] S. Ulmer et al. “A cryogenic detection system at 28.9 {MHZ} for the non-destructive observation of a single proton at low particle energy,” in: *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 705 (2013), pp. 55–60.
- [Ulm15] S. Ulmer et al. “High-precision comparison of the antiproton-to-proton charge-to-mass ratio,” in: *Nature* 524.7564 (2015). Letter, pp. 196–199.
- [WA07] R. L Wong and I. J. Amster. “Experimental evidence for space-charge effects between ions of the same mass-to-charge in Fourier-transform ion cyclotron resonance mass spectrometry,” in: *International journal of mass spectrometry* 265.2 (2007), pp. 99–105.

- [Wan12] M. Wang et al. “The AME2012 atomic mass evaluation,” in: *CPC(HEP & NP)* 36 (2012), pp. 1603–2014.
- [Wan17] M. Wang et al. “The AME2016 atomic mass evaluation (II). Tables, graphs and references,” in: *Chinese Physics C* 41.3 (2017), p. 030003.
- [Wap58] A. H. Wapstra. *Handbuch d. Physik*. Vol. 38. 1. Berlin - Gottingen - Heidelberg, 1958.
- [Web05] C. Weber et al. “Weighing excited nuclear states with a Penning trap mass spectrometer,” in: *Physics Letters A* 347.1–3 (2005). Einstein Special IssueSpecial Issue in celebration of this year’s World of Physics and the centenary of Einstein’s annus mirabilisEinstein Special Issue, pp. 81–87.
- [Wei35] C. F. V. Weizsäcker. “Zur Theorie der Kernmassen,” in: *Zeitschrift für Physik* 96 (July 1935), pp. 431–458.
- [Wel17a] A. Welker et al. “Binding Energy of ^{79}Cu : Probing the Structure of the Doubly Magic ^{78}Ni from Only One Proton Away,” in: *Phys. Rev. Lett.* 119 (19 2017), p. 192502.
- [Wel17b] A. Welker et al. “Precision electron-capture energy in ^{202}Pb and its relevance for neutrino mass determination,” in: *The European Physical Journal A* 53.7 (2017), p. 153.
- [Wid28] R. Wideröe. “Über ein neues Prinzip zur Herstellung hoher Spannungen,” in: *Archiv für Elektrotechnik* 21.4 (1928), pp. 387–406.
- [Wie13] F. Wienholtz et al. “Masses of exotic calcium isotopes pin down nuclear forces,” in: *Nature* 498.7454 (2013). Letter, pp. 346–349.
- [Wie17] F. Wienholtz et al. “Mass-selective ion ejection from multi-reflection time-of-flight devices via the pulsed in-trap lift,” in: *Int. J. Mass Spectrom.* (2017).
- [Wie98] W. Wien. “Untersuchungen über die electrische Entladung in verdünnten Gasen,” in: *Annalen der Physik* 301 (1898), pp. 440–452.
- [Win90] J. A. Winger et al. “Structure of ^{76}Zn from ^{76}Cu decay and systematics of neutron-rich Zn nuclei,” in: *Phys. Rev. C* 42 (3 1990), pp. 954–960.
- [Wiz79] J. L. Wiza. “Microchannel plate detectors,” in: *Nuclear Instruments and Methods* 162.1-3 (1979), pp. 587–601.
- [WM55] W. C. Wiley and I. H. McLaren. “Time [U+2010] of [U+2010] Flight Mass Spectrometer with Improved Resolution,” in: *Review of Scientific Instruments* 26.12 (1955), pp. 1150–1157.
- [Wol11] R. N. Wolf. “A multi-reflection time-of-flight mass separator for isobaric purification of radioactive ion beams,” in: *Hyperfine Interactions* 199.1 (2011), pp. 115–122.

- [Wol12] R. N. Wolf et al. “On-line separation of short-lived nuclei by a multi-reflection time-of-flight device,” in: *Nucl. Instr. and Meth. A* 686 (2012), pp. 82–90.
- [Wol13a] R. N. Wolf et al. “Plumbing Neutron Stars to New Depths with the Binding Energy of the Exotic Nuclide ^{82}Zn ,” in: *Phys. Rev. Lett.* 110 (4 2013), p. 041101.
- [Wol13b] R.N. Wolf et al. “ISOLTRAP’s multi-reflection time-of-flight mass separator/spectrometer,” in: *Int. J. Mass Spectrom.* 349-350.0 (2013). 100 years of Mass Spectrometry, pp. 123 –133.
- [WP90] H. Wollnik and M. Przewloka. “Time-of-flight mass spectrometers with multiply reflected ion trajectories,” in: *International Journal of Mass Spectrometry and Ion Processes* 96.3 (1990), pp. 267 –274.
- [WS54] R. D. Woods and D. S. Saxon. “Diffuse Surface Optical Model for Nucleon-Nuclei Scattering,” in: *Phys. Rev.* 95 (2 1954), pp. 577–578.
- [WW15] F. Wienholtz, A. Welker, et al. “Towards ultrahigh-resolution multi-reflection time-of-flight mass spectrometry at ISOLTRAP,” in: *Physica Scripta* 2015.T166 (2015), p. 014068.
- [WW16] R.N. Wolf, A. Welker, et al. “Background-free beta-decay half-life measurements by in-trap decay and high-resolution MR-ToF mass analysis,” in: *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 376 (2016), pp. 275 –280.
- [Xu14] Z. Y. Xu et al. “ β -Decay Half-Lives of $^{76,77}\text{Co}$, $^{79,80}\text{Ni}$, and ^{81}Cu : Experimental Indication of a Doubly Magic ^{78}Ni ,” in: *Phys. Rev. Lett.* 113 (3 2014), p. 032505.
- [Yan16] X. F. Yang et al. “Isomer Shift and Magnetic Moment of the Long-Lived $1/2^+$ Isomer in $^{79}_{30}\text{Zn}_{49}$: Signature of Shape Coexistence near ^{78}Ni ,” in: *Phys. Rev. Lett.* 116 (18 2016), p. 182502.
- [Yas86] S. Yasumi et al. “The mass of the electron neutrino using electron capture in ^{163}Ho ,” in: *Physics Letters B* 181.1 (1986), pp. 169 –172.
- [Yor13] D. T. Yordanov et al. “Spins, Electromagnetic Moments, and Isomers of Cd 107- 129,” in: *Physical review letters* 110.19 (2013), p. 192501.
- [Zhu08] S. Zhu et al. In: *Atom. D. and Nucl. D. Tabl.* 109 (2008), p. 699.
- [Zub01] K. Zuber. “COBRA—double beta decay searches using CdTe detectors,” in: *Physics Letters B* 519.1–2 (2001), pp. 1 –7.

Acknowledgment

I would like to start by thanking my parents Andrea and Fred Welker, who have always offered me constructive support throughout my life and who have made everything possible. In addition to my parents, I would like to thank my four brothers René, Marcel, Leon and Jean-Pierre, for the fruitful and constructive discussions in my life. During the doctoral thesis phase and beyond, my beloved partner Karmen Džeko has helped me most of all with her wise and encouraging words, for which I am more than grateful.

I would thank Prof. Dr. Kai Zuber my PhD supervisor and Prof. Dr. Klaus Blaum as my second supervisor for the offer and the opportunity to work at ISOLTRAP. Thank you both for being available whenever advice was needed.

I would also like to thank the former local ISOLTRAP spokes person Dr. Susanne Kreim for the introduction at the beginning of my time at ISOLTRAP and the constructive words during the correction of some manuscripts. In this context, I would say a big thank to all current ISOLTRAPers who have helped me during my doctoral thesis time. The great team work during beam times in connection with plenty of discussions. I would like to thank Frank Wienholtz for his helpful words on hardware issues. My thanks go to Dr. Dinko Atanasov and Maxime Mougeot for software related questions concerning the LabView code of ISOLTRAP's control system. I would like to thank Jonas Karthein for his energetic support with his event sorting algorithm and for his helpful Python codes. Last but not least, I would really like to thank Dr. Vladimir Manea, the current local ISOLTRAP spokes person, who helped me the most in writing papers, as well as constructive and in-depth discussions with which he has supported me with his profound knowledge. I really appreciate all your efforts. Additionally, I would also like to thank Dr. Vladimir Manea and Dr. Magdalena Kowalska during the correction phase of this thesis. Furthermore, I thank Prof. Dr. David Lunney for his ideas and advice for the copper article and Dr. Marco Rosenbusch for his parallel cross-check evaluation of the copper data. Last but not least, I would like to thank Dr. Sergey Eliseev and Pavel Filianin for their help during the analysis phase of the thallium article.

I would like to express my thanks for the support of the Wolfgang Gentner scholarship, the graduate college mass, spectrum, symmetry (GK 1504) of the Humboldt university of excellence Berlin and the graduate academy of the TU Dresden of excellence.

Versicherung

Hiermit versichere ich, Andree Welker, geboren am 13. Februar 1986 in Bad Kreuznach, dass ich die vorliegende Arbeit ohne unzulässige Hilfe Dritter und ohne Benutzung anderer als der angegebenen Hilfsmittel angefertigt habe; die aus fremden Quellen direkt oder indirekt übernommenen Gedanken sind als solche kenntlich gemacht. Jedes Bild, Skizze oder Simulation, welche nicht extra eines anderen Ursprungs ausgewiesen wurde, unterliegt dem Urheberrechtsschutz dieser Arbeit. Die Arbeit wurde bisher weder im Inland noch im Ausland in gleicher oder ähnlicher Form einer anderen Prüfungsbehörde vorgelegt.

Die Dissertation wurde an der Technischen Universität Dresden sowie am Forschungszentrum CERN in Genf unter der Betreuung von Prof. Dr. Kai Zuber angefertigt.

Es wurden zuvor keine Promotionsvorhaben unternommen. Ich erkenne die Promotionsordnung der Fakultät Mathematik und Naturwissenschaften der TU Dresden vom 23.02.2011 an.

Genf den, 14. März 2018