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Studies and Cooling of Highly Charged Ion Ensembles in the ARTEMIS Trap and High-Precision Mass Measurements of ²²¹Fr, ²¹⁹Rn, ²¹³Bi, ²¹¹Pb, ²⁰⁹Pb, ²⁰⁷Tl and ²⁰⁷Pb at SHIPTRAP

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Zusammenfassung

Penning-Fallen eröffnen einzigartige experimentelle Möglichkeiten für Massenspektrometrie und Spektroskopie atomarer Ionen mit hoher Präzision. Zwei derartige Experimente auf Basis von Penning-Fallen sind SHIPTRAP und ARTEMIS am GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt.

Das ARTEMIS-Experiment dient der Messung des magnetischen Moments (g-Faktors) des Elektrons in schweren, hoch geladenen Ionen auf dem Niveau von 10^{-9} mittels einer Laser-Mikrowellen-Doppelresonanzspektroskopie. Derartige Messungen stellen hoch-stringente Tests der QED in extremen Feldern dar. Das Ion der Wahl für die Demonstration und Entwicklung der experimentellen Methoden ist 40 Ar¹³⁺, welches intern in der Falle produziert werden kann, für spätere Messungen ist 209 Bi⁸²⁺ vorgesehen. Für die Messungen ist jeweils die Präparation einer sortenreinen und gekühlten Wolke von Ionen in der Spektroskopie-Falle notwendig. Im Rahmen dieser Arbeit wurde das System für den zerstörungsfrei Nachweis und die Kühlung der Ionen optimiert, und Produktion, Transport, Kühlung, Selektion und Speicherung der Ar¹³⁺-Ionen wurden systematisch demonstriert.

An SHIPTRAP wurden Präzisions-Messungen der Massen von Tochternukliden der langlebigen Radio-Nuklide ²²⁵Ac und ²²³Ra durchgeführt, und zwar mittels einer phasenempfindlichen Ionenzyklotron-Resonanz-Methode. Die Massen von ²²¹Fr, ²¹⁹Rn, ²¹³Bi, ²¹¹Pb, ²⁰⁹Pb, ²⁰⁷Tl und ²⁰⁷Pb wurden mit einer relativen Genauigkeit von 10^{-9} gemessen und erlauben eine Steigerung der Genauigkeit anderer Massen in dieser Region. Einige dieser Massen finden direkten Eingang in die *g*-Faktor-Messungen, etwa im Fall von ²⁰⁹Bi⁸²⁺, welches von Interesse für ARTEMIS ist. Außerdem ist das Dublett ²⁰⁵Tl / ²⁰⁵Pb, das in der nuklearen Astrophysik von großer Bedeutung ist, ebenfalls mit den gemessenen Massen verbunden.

Abstract

Penning traps open up unique experimental possibilities for mass spectrometry and spectroscopy of atomic ions with high precision. Two such experiments based on Penning traps are SHIPTRAP and ARTEMIS at the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt.

The ARTEMIS experiment is designed to measure the magnetic moment (g-factor) of an electron in heavy, highly charged ions at the 10^{-9} level, by the means of lasermicrowave double-resonance spectroscopy. Such measurements represent highly stringent tests of QED in extreme fields. The ion of choice for the demonstration and development of the experimental methods is ${}^{40}\text{Ar}{}^{13+}$, which can be produced internally in the trap, for later measurements ${}^{209}\text{Bi}{}^{82+}$ is foreseen. For each of the measurements, the preparation of a cleaned and cooled cloud of ions in the spectroscopy trap is necessary. In this work, the system is optimised for non-destructive detection and cooling of the ions, and production, transport, cooling, selection and storage of the Ar¹³⁺ ions are systematically demonstrated.

Precision mass measurements of the long-lived decay products of 225 Ac and 223 Ra are carried out at SHIPTRAP, using the phase-imaging ion-cyclotron-resonance technique. The masses of 221 Fr, 219 Rn, 213 Bi, 211 Pb, 209 Pb, 207 Tl and 207 Pb are measured with a relative precision of 10^{-9} , allowing an increase in the accuracy of other masses in this region. Some of these masses find direct input into the *g*-factor measurements, such as in the case of 209 Bi, which is of interest to ARTEMIS. Furthermore, the doublet 205 Tl / 205 Pb, which is of great significance in nuclear astrophysics, is also linked to the measured masses.

"Zum Mitnehmen oder hier essen?" average graduate student

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Chapter 1 Introduction

According to the Standard Model of particle physics, there are three fundamental forces by which elementary particles interact: electromagnetic, strong and weak force. In addition, all particles interact through gravity. A physical theory corresponds to each of the forces such as the theory of quantum electrodynamics (QED) for the electromagnetic force [1, 2], the quantum theory of chromodynamics (QCD) for the strong force [3, 4], the Glashow-Weinberg-Salam (GWS) theory¹ for the weak force [5] and the classical theory of gravity with relativistic generalisation in the Einstein's general theory of relativity [6]. The theory of quantum electrodynamics describes the fundamental interaction between the photons and charged particles. It is one of the most successful and most tested theories of modern physics. A graphical way of representing the mathematical expressions that describe the QED interactions was developed by Richard Feynman in 1948 in the form of Feynman diagrams [1]. Each vertex in the Feynman diagram contributes $\sqrt{\alpha}$, where α is the coupling constant (fine-structure constant) that quantifies the interaction strength in QED. Since the value of this interaction parameter is small ($\alpha \approx 1/137$), for the light mass systems $(Z\alpha \ll 1)$, the QED effects can be explored perturbatively by expansions in α using the Feynman diagrams.

High-precision measurements are a powerful tool to test the Standard Model (SM) and search for physics beyond the standard model (BSM). The magnetic moment of an electron is one of the most precisely measured quantities of an elementary particle and provides a stringent test for the theory of quantum electrodynamics. The most recent measurement for the free electron's magnetic moment is performed in the group of G. Gabrielse in 2022 with an uncertainty of 1.3×10^{-13} [7]. These measurements are consistent with and 2.2 times more precise than the previous measurements from

 $^{^{1}}$ The GWS theory considers the electromagnetic and weak interaction as different actualisations of a single electroweak theory.

the same group in 2008 [8]. These high-precision measurements allow testing QED and make it one of the best tested theories in physics.

An extension to the theory of quantum electrodynamics is the bound-state quantum electrodynamics theory in the extreme electric and magnetic fields of the nucleus. In the case of heavy, highly charged ions such as $^{208}\text{Pb}^{81+}$ and $^{209}\text{Bi}^{82+}$, the fields are as high as $E_{nuc} \approx 10^{16} \text{Vcm}^{-1}$ and $B_{nuc} \approx 10^7 \text{ T}$ [9, 10]. These high fields cannot be generated in the laboratories. Furthermore, there is a strong scaling with the atomic number Z for the fine- (Z⁴) and hyperfine-structure (Z³) transitions. These transitions are in the ultraviolet or optical region of the spectrum which is easily accessible through laser spectroscopy. Therefore, the heavy, highly charged ions provide perfect testing environments for the theory of bound-state quantum electrodynamics. Apart from the bound-state QED effects, the magnetic moment measurements in these heavy few-electron systems also provide an insight into many other properties such as the nuclear structure and size, relativistic electron correlation and higher-order Zeeman effects.

In contrast to the electromagnetic force, the strong interaction has a large coupling constant. Hence, the perturbative treatment cannot be applied in this case, as for the quantum theory of electrodynamics. The strong force, which is mediated by gluons, is short range and is unfortunately not as well understood as the electromagnetic force. Therefore, several nuclear models have been developed over the years to determine the binding energy of the nuclei. The binding energy of a nucleus carries information about its structure and can readily be obtained from direct mass measurements [11]. Nuclear models have comparably low predictive power. In order to constrain some of the free parameters in these models, it would be desirable to have the mass measurements of as many nuclei as possible with the highest possible precision.

Penning traps are excellent tools for high-precision spectroscopy and mass spectrometry experiments. The ARTEMIS experiment at GSI Helmholtz Center for Heavy Ion Research, Germany, aims to perform high-precision measurements for the magnetic moment of an electron bound in a heavy, highly charged ion using the laser-microwave double-resonance spectroscopy technique [12]. In order to achieve this milestone, a pure cooled cloud of highly charged ions needs to be confined in a Penning trap set-up in a cryogenic environment under ultra-high vacuum conditions better than 10^{-14} mbar. The cyclotron frequency of the ions is measured using non-destructive electronic detectors through the amplification of image currents induced on the electrodes. Contrary to the conventional continuous Stern-Gerlach scheme which is based on the measurement of spin flips in other g-factor experiments [13, 14, 15, 16], the ARTEMIS experiment is designed to use the laser-microwave double-resonance technique for measurement of the Larmor frequency. The technique probes the Zeeman sub-states using a tunable microwave radiation for a closed fine structure or hyperfine structure transition which is driven by a laser. A distinct feature of the double-resonance technique arises from the ability to measure a) the g-factor in the ions with non-zero nuclear spins [12] and b) the bound electron magnetic moment and nuclear magnetic moment simultaneously. A novel half-open Penning trap design [17] has been implemented in the set-up to maximise the optical access for laser spectroscopy of the ions. The first experiment at ARTEMIS aims to measure the g-factor of 40 Ar¹³⁺ with a 10⁻⁹ level of precision [18]. The experiment is currently in the commissioning phase.

This work presents the preliminary tests on ${}^{40}\text{Ar}^{13+}$ in order to prepare a cooled pure cloud in the ARTEMIS Penning trap. In addition to this, upgrades to the nondestructive detection systems performed at the ARTEMIS set-up are discussed. Highprecision measurements of the magnetic moment of hydrogen-like bismuth, ${}^{209}\text{Bi}{}^{82+}$, are foreseen in the upcoming beamtime in 2024. The mass of ${}^{209}\text{Bi}$ is an external input parameter and affects the final achievable precision of the measurement. Thus, the precise knowledge of its mass value is essential to the experiment.

A Penning trap experiment at GSI which performs high-precision mass measurements is the SHIPTRAP mass spectrometer. The mass of an atom is one of its basic properties and provides information about its constituents and their interactions. High-precision mass measurements are of great importance to probe the nuclear structure $(\delta m/m \approx 10^{-6} - 10^{-8})$ and to support astrophysical models of nucleosynthesis $(\delta m/m \approx 10^{-7})$ [19]. The SHIPTRAP Penning trap experiment uses the Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique to measure the cyclotron frequency of the ion of interest relative to a reference ion species [20]. This destructive detection technique enables measurements with short-lived radio-nuclides having half-lives well below one second [20, 21] with a precision of $\delta m/m \approx 10^{-9}$. Using this technique, the masses of seven radio-nuclides (²²¹Fr, ²¹⁹Rn, ²¹³Bi, ²¹¹Pb, $^{209}\mathrm{Pb},\,^{207}\mathrm{Tl}$ and $^{207}\mathrm{Pb})$ from the decay chains of $^{225}\mathrm{Ac}$ and $^{223}\mathrm{Ra}$ have been measured within the scope of this thesis. Since the mass of 209 Bi is linked to the mass of 209 Pb through beta decay, such a measurement can improve the present uncertainty in the mass of ²⁰⁹Bi. Furthermore, the ²⁰⁵Tl and ²⁰⁵Pb doublet is of great significance in nuclear astrophysics as an s-process (slow-neutron capture process) cosmochronometer [22] and also to determine the nuclear matrix elements related to the doublet at the LOREX project (LORandite EXperiment) [23, 24]. This doublet is related to 207 Tl and 207 Pb through (t, p) and (n, γ) nuclear reactions. Therefore, the 205 Tl / 205 Pb doublet masses are linked to the mass measurements performed in this work.

This thesis is structured into eight chapters. Chapter 2 discusses the fundamentals of the most essential tool within the framework of the high-precision measurements at the SHIPTRAP and the ARTEMIS experiment: the Penning trap. The techniques for ion manipulation, cooling and detection are also discussed in this chapter. In chapter 3, theoretical motivations leading to the ARTEMIS experimental set-up and for the mass measurements conducted at the SHIPTRAP mass spectrometer are described along with the principle of measurements utilised in both of these experiments. The experimental set-up for the ARTEMIS Penning trap along with the important upgrades on the non-destructive detection system are reported in chapter 4. Afterwards, in chapter 5, the investigations performed on the ion clouds confined in the ARTEMIS experiment together with an estimation of the electronic load capacitance of the trap and characterisation of the cooling and cleaning techniques for Ar^{13+} ions are discussed. Chapter 6 gives an overview of the experimental set-up of the SHIPTRAP mass spectrometer. The mass measurements of the radio-nuclides obtained from the recoil-ion sources ²²⁵Ac and ²²³Ra installed in the cryogenic gas cell of the SHIPTRAP experiment are discussed in chapter 7, followed by the conclusions and outlook for both of the Penning trap experiments in chapter 8.

Chapter 2

Penning trap fundamentals and techniques

According to Earnshaw's theorem, it is impossible to confine a charged particle in all three dimensions using electrostatic field only [25]. Thus, in order to have a stable confinement of particles using electrostatic field, an additional confining field is applied, such as radio-frequency (rf) field in the case of Paul trap [26] or a homogeneous magnetostatic field in a Penning trap [27]. The homogeneous axial magnetostatic field in a Penning trap provides the radial confinement and the electrostatic field confines the particles axially. The Penning trap is the most widely used tool for highprecision experiments such as mass spectrometry and q-factor measurements. The first concept of Penning trap was already introduced in 1939 by F. M. Penning when he used the magnetic field to improve the operation of vacuum gauge [28]. Later in 1949, J. R. Pierce pointed out in his book 'Theory and Design of Electron Beams' [29] that it is possible to have sinusoidal oscillations of electrons in the presence of harmonic electric and axial magnetic field. This inspired Hans G. Dehmelt to build such a device which uses static electric and magnetic fields to store electrons, which he named as 'Penning trap' after Frans Michel Penning. In 1989, H. G. Dehmelt was awarded the Nobel Prize together with Wolfgang Paul, for 'the development of the ion trap technique' and they shared the other half of the prize with Norman F. Ramsey 'for the invention of the separated oscillatory fields method and its use in the hydrogen maser and other atomic clocks'.

There are several advantages of Penning traps over the other trapping devices such as optical traps, acoustic traps or Paul traps. Penning traps a) can confine ions with a wide range of charge-to-mass ratios simultaneously, b) have tighter confinements which allows higher energy particles to be trapped, c) have no inherent heating due to only static fields only, and d) allow radio-frequency based ion cloud manipulations. However, the strong confining magnetic fields are produced using superconducting magnets which lead to non-compact experimental set-ups.

In this chapter, different types of Penning trap designs utilised at ARTEMIS and SHIPTRAP are discussed. A detailed discussion for the ion motion and manipulation in the Penning trap along with the frequency shifts in the ion motions and ion cooling and detection techniques is also presented.

2.1 The principal design

A set of three electrodes with a central ring electrode and two endcap electrodes, on which the voltages can be applied in such a way as to have a harmonic potential well, and an axial magnetic field, constitutes an elementary Penning trap. The electric field confines the particles axially while the magnetic field forces the charged particles in a circular motion along a perpendicular plane (x-y plane).

The oldest Penning trap geometry is the hyperbolic trap with the hyperbolic ring electrode and two endcap electrodes. This trap geometry has an advantage that for any set of voltages applied to the electrodes, quadrupole potential well is created which causes harmonic axial oscillation of the charged particles [30].

Although a precisely machined hyperbolic trap creates a perfectly harmonic trap, yet there are some limitations, such as the lack of adequate optical and mechanical access. The introduction of slits along the axial and azimuthal directions leads to non-zero octupolar components (dominant electric imperfection term) which distort the quadrupolar potential leading to anharmonicities. At the time of first use of hyperbolic trap, it was difficult to have precisely machined hyperbolic electrodes, but this has been greatly improved over time with advancements in machining techniques.

In order to overcome the challenges faced in hyperbolic trap, an alternative is the use of hollow cylindrical ring electrodes with two flat endcap electrodes (figure 2.1). This geometry has a cylindrical symmetry along the z-axis and a mirror symmetry along the x-y plane.

2.1.1 Mechanical compensation

In case of a cylindrical trap, the deviation from the hyperbolic shape leads to nonquadrupolar potential at the trap center and hence the axial confinement is not harmonic in general. With the correct choice of the ratio ρ_0/z_0 , where ρ_0 is the inner radius of the hollow cylindrical trap electrode and z_0 is the distance of the endcap



Figure 2.1: Schematic of the mechanically compensated cylindrical Penning trap with open and closed endcaps. ρ_0 and z_0 are distance of the ring and either endcap from center of the trap. U_r and U_e are, respectively, the potentials on the ring and the endcap electrodes.

from the center (figure 2.1), a harmonic potential well can be created at the trap center. This ratio for a closed cylindrical trap is:

$$\frac{\rho_0}{z_0} = 1.203 \tag{2.1}$$

For this value, the potential at the trap center becomes quadrupolar. This is called mechanical compensation or geometric compensation [31] as shown in figure 2.1.

However, the closed endcap still does not provide with an excellent optical and ion loading access, thus requiring further modifications in the geometry in the form of open endcap design or half-open endcap design, as discussed in section 2.1.3.

2.1.2 Electrical compensation

The cylindrical trap can also be electrically compensated to provide a quadrupolar potential at the trap center by adding a set of electrodes (called the 'compensation electrodes') on either side of the ring electrode. The potential at the compensation electrodes is applied in such a way that a harmonic potential well is created at the center. This provides another degree of freedom to make the trap harmonic, with a tunable voltage U_c at the compensation electrodes. Thus, this represents an electrically compensated trap [32]. For a cylindrical trap, the electrostatic potential near the trap center can be written as:

$$U = \frac{U_0}{2} \sum_{k=0}^{\infty} C_k \left(\frac{r}{d}\right)^k P_k(\cos\theta)$$
(2.2)

where $U_0 = U_r - U_e$ is the voltage applied between ring electrode and the endcap electrodes which axially confines the particles, $r = \sqrt{\rho^2 + z^2}$ is the distance of the particles to the trap center with ρ and z being the radial and axial distance, respectively, and $P_k(\cos \theta)$ is the k-th order Legendre polynomial with

$$\cos\theta = \frac{z}{\sqrt{\rho^2 + z^2}} = \frac{z}{r},\tag{2.3}$$

d is the 'characteristic trap dimension' given by:

$$d^2 = \frac{z_0^2}{2} + \frac{\rho_0^2}{4}.$$
 (2.4)

Since the trap has cylindrical symmetry with respect to the z-axis and mirror symmetry at trap center (z=0) along the perpendicular plane (x-y plane), the C_k coefficients are zero for odd values of k. The coefficients C_k for even k are defined as:

- C_0 : overall potential offset.
- C_2 : quadrupolar term, represents the efficiency of the trap to create potential wells from applied voltages; determines the potential at the trap center and hence the exact axial oscillation frequency of charged particles.
- C_4 and C_6 : octupole and dodecapole components, characterize the electric imperfections in the trap.

For a hyperbolic Penning trap, $C_2 = 1$ whereas for a cylindrical trap $C_2 = 0.5$. C_4 characterises as the leading order electric imperfection term. The terms above C_6 can generally be neglected on account of their small contribution (less than the experimental resolution). Since C_4 and C_6 contribute to anharmonicities in the trapping potential, they should be kept to a minimum value. For an electrically compensated trap, C_2 and C_4 can be written as:

$$C_2 = C_2^{(0)} + D_2 \frac{U_c}{U_o}$$
 and $C_4 = C_4^{(0)} + D_4 \frac{U_c}{U_0}$ (2.5)

where $C_2^{(0)}, C_4^{(0)}$ and D_2, D_4 are given by trap geometry [32] and U_c is the voltage on the compensation electrodes. The ratio of these two voltages U_c/U_0 is called the 'tuning ratio' and can be chosen such that $C_4 = 0$. Since, this tuning ratio also appears in C_2 , thus for a non-zero D_2 any change in U_c/U_0 affects the value of C_2 . This in turn changes the oscillation frequency of the ions. However, for any given z_c/z_0 , there is a particular value of ρ_0/z_0 such that $D_2 = 0$. Such a trap is called **orthogonal trap**. For a certain geometric condition given by:

$$\frac{z_c}{z_0} = 0.8351$$
 and $\frac{\rho_0}{z_0} = 1.0239$

both C_4 and C_6 are zero and the trap is harmonic.



Figure 2.2: Schematic of an electrically compensated open-endcap Penning trap showing radial and axial confinement. The presence of an extra set of electrodes on either side of the ring electrode (compensation electrodes with a potential U_c) provides additional degree of freedom to make the trap harmonic. Here, z_e is the height of the endcap electrode and z_c is the length of the compensation electrode.

2.1.3 Half-open endcap design

In order to mimic the potential of a closed endcap in an open-endcap Penning trap design, elongated endcap electrodes are used on either end of the electrode stack [33]. The ions confined at the ring electrode are thus very far from the trap opening which limits the opening angle of light cone from the trap center and subsequently limits the optical access to the ions. A possible design variation is the so-called 'half-open' design where one closed endcap is replaced by a set of open cylindrical electrodes ('anti-compensator' and 'anti-ring' forming the 'anti-trap') and the other one is replaced by a closed endcap which is electrically closed and optically transparent (e.g. a conducting mesh or a conductive coating on a transparent window) [34]. One such trap has been implemented at the ARTEMIS experiment (section 4.2.2). The open electrode stack allows the injection and ejection of particles while the optically transparent window provides for larger light collection efficiency. Because the ions are closer to the window, they have greater opening angle for the light cone. As a result, the light collection efficiency significantly increases and the solid angle of detection becomes about 11 times the value in an open-endcap design [35].

2. Penning trap fundamentals and techniques



Figure 2.3: Schematic of the open-endcap and half-open Penning trap design. It can be seen that replacement of an open endcap with a closed transparent conduction window increases the solid angle by ten folds. Also, the other endcap electrode has been replaced with anti-trap electrodes, as discussed in the text.

In ARTEMIS, the trap geometry is a combination of many of these concepts forming a double trap with a mechanically compensated creation trap and an electrically compensated half-open spectroscopy trap (section 4.2).

2.2 Ion motion in a Penning trap

A charged particle confined in superposition of electrostatic field, $\vec{E} = -\nabla U$, and axial magnetic field, $\vec{B} = B_0 \hat{z}$, in a Penning trap undergoes three independent oscillatory motions. Under the effect of a quadrupolar potential U, given by:

$$U \propto 2z^2 - x^2 - y^2 \tag{2.6}$$

the particle undergoes harmonic axial oscillations with frequency ν_z in the z-direction and experiences a repulsive electrostatic force in x and y directions. A uniform magnetic field in the z direction forces the charged particle in a circular motion around the magnetic field axis with a frequency ν_c . In a Penning trap, since the particle is in a crossed electric and magnetic field, the particle undergoes a drift motion with frequency ν_- . The presence of electric field modifies the cyclotron motion to have a



Figure 2.4: Motion of an ion in a Penning trap. The particle undergoes three oscillatory motions, the axial oscillation along z-axis with frequency ν_z and two radial motions in the x-y plane with frequencies ν_+ (modified cyclotron motion) and ν_- (magnetron motion).

frequency ν_+ instead of ν_c . The trajectory of a particle followed in a Penning trap resembles a crown made from a hollow cylindrical spring (figure 2.4) and the projection in x-y plane is an epitrochoid.

A particle of charge q and mass m in a Penning trap experiences Lorentz force:

$$\vec{F} = q(-\nabla U + \vec{v} \times \vec{B}) \tag{2.7}$$

which gives rise to a simple harmonic oscillation of the particle in the axial direction with a frequency ν_z :

$$\nu_z = \frac{1}{2\pi} \sqrt{\frac{qC_2 U_0}{md^2}}$$
(2.8)

where U_0 is the applied trap potential, d is the 'characteristic trap dimension' (equation 2.4) and C_2 is the trap coefficient as defined in section 2.1. The free cyclotron motion frequency of an ion in the presence of only a homogeneous magnetic field is given by:

$$\nu_c = \frac{qB_0}{2\pi m} \tag{2.9}$$

As discussed already, the presence of electric field in a Penning trap modifies the cyclotron motion to have a frequency ν_+ known as the 'reduced cyclotron frequency'. Since the cyclotron motion drifts along electric equipotential lines, the center of cyclotron motion moves slowly about the central axis of the Penning trap, with a frequency ν_- . This radial oscillation is the magnetron motion of an ion. These radial

motional frequencies are given by:

$$\nu_{+} = \frac{\nu_{c}}{2} + \sqrt{\frac{\nu_{c}^{2}}{4} - \frac{\nu_{z}^{2}}{2}}$$
 and $\nu_{-} = \frac{\nu_{c}}{2} - \sqrt{\frac{\nu_{c}^{2}}{4} - \frac{\nu_{z}^{2}}{2}}$ (2.10)

The detailed calculations of these three eigenfrequencies of ion motion can be found in various text books. On Taylor expansion of the magnetron frequency ν_- , it can be observed that the magnetron frequency is independent of q/m of the ion to first order.

$$\nu_{-} \approx \frac{C_2 U_0}{4\pi d^2 B_0} \tag{2.11}$$

Measurement of cyclotron frequency is one of the key tools in high-precision Penning trap experiments such as ARTEMIS and SHIPTRAP. In the case of highprecision mass spectrometry experiments such as SHIPTRAP, the true cyclotron frequency is measured using the sideband method:

$$\nu_c \approx \nu_+ + \nu_- \tag{2.12}$$

This relation also holds for an ideal Penning trap in the absence of any electric or magnetic field imperfections.¹ However in a real trap, this relation is only an approximation since it is impossible to avoid the imperfections which creep in such as electric field fluctuations, patch potentials or imperfect machining of trap electrodes. For the experiments which aim for higher orders of precision, such as ARTEMIS (aiming for 10^{-9} precision), the determination of ν_c is based on Brown-Gabrielse invariance theorem [36]:

$$\nu_c^2 = \nu_z^2 + \nu_+^2 + \nu_-^2 \tag{2.13}$$

 ν_c remains unaffected by the first order misalignments and ellipticities. Although these alter the individual eigenfrequencies, yet the effects get canceled in the calculation of ν_c .

The ion motion usually follows the hierarchy $\nu_c > \nu_+ >> \nu_z >> \nu_-$. For an ideal trap with a single confined charged particle, these eigenfrequencies are independent of each other. A change in the trapping voltage modifies the axial motional frequency. In addition to ν_z , the radial frequencies also, to an extent, depend on the trapping potential. This is shown in figure 2.5. There exists a value of trapping voltage U', beyond which $\nu_z > \nu_+$. However, the Penning trap is not operated in this domain since the particles will no longer be trapped in all three dimensions, rather will escape

 $^{^{1}}$ Equation 2.12 is also used in the cases where the achievable precision is limited by other factors and, therefore, electric and magnetic field imperfections do not significantly influence the precision achieved.



Figure 2.5: Variation of the oscillation frequency of ion motion as a function of trapping potential. ν_c is constant since it is not dependent on the electric potential. ν_z increases with the square root of trapping potential U, as seen from equation 2.8, ν_+ and ν_- depend on U through their dependence on ν_z (equation 2.10). Hence ν_+ decreases with U while ν_- increases with increase in U. At U_{max} , the term under the square root becomes zero and $\nu_+ = \nu_- = \nu_c/2$ and beyond this potential, confinement is lost. Beyond a certain potential U', $\nu_+ < \nu_z$ and the particles are lost radially.

radially. It should be noted that the magnetic confinement in a trap is several orders of magnitude stronger than the electric confinement [35].

For the particle trajectory to be stable, the term under the square root in equation 2.10 is required to be positive. This introduces a stability criteria defined by the parameter, γ :

$$\gamma = \frac{\nu_c}{\sqrt{2}\nu_z} > 1 \implies \nu_c > \sqrt{2}\nu_z \implies B_0 > \sqrt{\frac{2mC_2U_0}{qd^2}}$$
(2.14)

Rather than the actual trajectory of the particle in a Penning trap, the observables such as oscillation frequency and amplitude of oscillation are of greater interest. The corresponding amplitudes for three different eigenmotions are given by:

$$a_{+}^{2} = \frac{8\pi^{2}E_{+}}{m(\nu_{+}^{2} - \nu_{z}^{2}/2)} \quad ; \quad a_{-}^{2} = \frac{8\pi^{2}E_{-}}{m(\nu_{-}^{2} - \nu_{z}^{2}/2)} \quad ; \quad a_{z}^{2} = \frac{4\pi^{2}E_{z}}{qC_{2}U_{0}}d^{2} \tag{2.15}$$

where E_+, E_- and E_z are the corresponding kinetic energies of motion. It should be noted that although the denominator for the amplitude square of magnetron motion becomes negative due to $\nu_z > \nu_-$, yet the right hand side of equation 2.15 is positive. This is because the kinetic energy of magnetron motion is also negative.

2.3 Frequency shifts in a Penning trap

Ions in a Penning trap undergo three oscillatory harmonic motions. In an ideal case, with no field imperfections and a single confined particle, these three motions are decoupled and independent of each other. However, it is impossible to have an ideal trap without any imperfections. The deviation from the idealised situation can be due to intrinsic properties of the trap such as the geometry of trap, imperfection in machining of electrodes, field misalignment and ellipticity, and temporal instability of electric and magnetic fields. In addition to these, the trapped particle effects such as image charge, image current and space charge effects can also lead to significant deviations. The latter becomes significant only in the case of confinement of multiple ions (ion clouds), as in the case of ARTEMIS. Since the ion clouds have an energy distribution, energy dependent effects also contribute to the frequency distribution. In order to avoid the energy dependent shifts, the confined ion clouds need to be thermalised using cooling procedures, as discussed in section 2.5.

2.3.1 Imperfections in confining fields

Anharmonicities in electrostatic fields and inhomogeneities of magnetosatic fields are the most prominent contributors to the imperfections in the trapping conditions. In a real trap, there are misalignment of the trap electrodes, deviations from the ideal geometry parameters (such as finite length endcap electrodes and segmented electrodes), mechanical imperfections and temporal variation of fields. These are some of the major sources of anharmonicities in electrostatic fields. The potential is never purely quadrupolar ($C_4 \neq 0$), thus leading to coupling between eigenmotions as well as dependence of motional frequencies on the average energies of motion E_+ , E_z and E_- (as can be seen in equation 2.17).

In addition to the spatial anharmonicities in the electrostatic potentials, the temporal instabilities cannot be overlooked. Once the thermal equilibrium is attained, the electrostatic fields can only be altered by changing the trapping potential (since the 'd' remains unchanged). The effect of atmospheric conditions on the output voltages from the high-precision voltage sources can be reduced by having a temperature controlled environment. 'Patch potentials' also contribute to the temporal anharmonicity of the electric field. Although a metallic surface is considered to be an electric equipotential surface, there are surface variation of potentials of up to several hundreds of millivolts over few micrometers [37]. These create local electrostatic fields due to different work

functions at different patches and can result in rf-noise on thermal fluctuations in patches.

The high magnetic fields in the Penning trap experiments are usually created by superconducting magnets. The fields are highly homogeneous at the trap center to about 10^{-7} in a region of $1 \, cm^3$. Just like the electrostatic potential, magnetic field can be written as [38]:

$$B_{z} = \sum_{k=0}^{\infty} B_{k} r^{k} P_{k}(\cos \theta) \text{ and } B_{\rho} = \sum_{k=0}^{\infty} B_{k} r^{k} \frac{1}{k+1} P_{k}'(\cos \theta)$$
(2.16)

where B_z , B_ρ are the axial and radial components of the magnetic field; $P_k(\cos \theta)$ is the Legendre polynomial and $P'_k(\cos \theta)$ is the first order associate Legendre polynomial [39]. B_0 (T) is the homogeneous field part, B_1 (T/m) is the linear gradient, and B_2 (T/m²) measures the strength of the magnetic bottle. A non-zero value of B_2 causes dependence of motional frequencies on the oscillation energies (see equation 2.17).

Ambient conditions such as pressure and temperature changes alter the helium evaporation rate which affect the internal temperatures. In the presence of strong magnetic fields, the materials with non-zero susceptibility can get magnetised and influence the magnetic field homogeneity. Additionally, the current in the superconducting coil of the magnet may also vary slowly over time due to 'flux-creep' phenomenon [40]. This occurs when the flux lines, which are pinned to the inhomogeneities of superconducting material, jump over pinning cites. The magnetic field fluctuations can be stabilised by having temperature stabilisation in magnet bore and pressure stabilisation in cryostat.

Thus, the shift observed due to imperfections in the confining electric and magnetic fields is given by [41]:

$$\begin{pmatrix} \Delta\nu_{+}/\nu_{+} \\ \Delta\nu_{z}/\nu_{z} \\ \Delta\nu_{-}/\nu_{-} \\ \Delta\nu_{L}/\nu_{L} \end{pmatrix} = (M_{E} + M_{B}) \begin{pmatrix} E_{+} \\ E_{z} \\ E_{-} \end{pmatrix}$$
(2.17)

where M_E and M_B are matrices defining the dependencies of oscillation frequencies on motional energies, respectively, due to electric and magnetic field imperfections.

$$M_E = \frac{6C_4}{qC_2U} \begin{pmatrix} \eta^4 & -\eta^2/2 & -\eta^2 \\ -\eta^2/2 & 1/4 & 1 \\ -\eta^2 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix} \quad \text{and} \quad M_B = \frac{2\pi^2}{m\nu_+\nu_-} \frac{B_2}{B} \begin{pmatrix} -\eta^2 & 1 & 2 \\ 1 & 0 & -1 \\ 2 & -1 & -2 \\ -\eta^2 & 1 & 2 \end{pmatrix}$$
(2.18)

and $\eta = \nu_z/\nu_+$, ν_L is the Larmor frequency (precession of particle's spin in magnetic field). Although ν_L is not a classical oscillation, it can be represented with the same formalism. Due to its purely magnetic property, bottom row in the matrix M_E is zero.

In order to reduce the effect of these imperfections on the free cyclotron frequency value, correction electrodes are incorporated into the trap design to diminish the electrostatic anharmonicities. It is observed that the invariance theorem holds true for electric imperfections but not for magnetic field shifts.

2.3.2 Field misalignment and ellipticity

Despite rigorous efforts, it is inevitable to have a tilt in the trap axis relative to the magnetic field axis. This causes coupling of motional frequencies and hence a shift in the free cyclotron frequency. The presence of additional radial quadrupolar components causes ellipticity in the electric field and breaks the rotational symmetry of the field. For ARTEMIS, the maximum possible value of ellipticity ϵ , is 0.0239. The electrostatic potential in the presence of a non-zero ϵ is [42]:

$$U = \frac{U_0}{2d^2} (2z^2 - x^2 - y^2 - \frac{\epsilon}{2}(x^2 - y^2))$$
(2.19)

Due to the precisely machined electrodes at ARTEMIS, ϵ has a small value. This leads to a small anharmonic contribution and hence can be neglected [43].

If θ is the polar angle representing the tilt between the trap axis and magnetic field axis, the frequency shifts are [36]

$$\nu'_z \approx \nu_z \left(1 - \frac{1}{4} (3+\epsilon) \sin^2 \theta \right) \quad \text{and} \quad \nu'_\pm \approx \nu_\pm + \frac{1}{2} \nu_- (3+\epsilon) \sin^2 \theta$$
 (2.20)

The shift in the free cyclotron frequency is given by [44]

$$\Delta \nu_c \approx \frac{9}{4} \nu_- \sin^2 \theta \tag{2.21}$$

For the cyclotron frequency measurement in SHIPTRAP, using the PI-ICR technique (section 3.6), inclination of the detector relative to the trap axis distorts the orbitimage projection on the detector. This alters the image to be elliptical and hence measures the phase angle ϕ^* instead of ϕ . Thus, the frequency shift is

$$\Delta \nu = \frac{\phi - \phi^*}{2\pi t_{acc}}.\tag{2.22}$$

Trap misalignment and ellipticity are unavoidable in a real Penning trap. Nevertheless, Brown-Gabrielse invariance theorem still remains valid such that $\nu_c^2 \approx \nu_+^{\prime 2} + \nu_z^{\prime 2} + \nu_-^{\prime 2}$.

2.3.3 Space charge effect

In many Penning trap experiments, there is usually more than one confined charged particle in the trap. Presence of different ion species leads to the cyclotron frequency shift. This is due to interaction of their center-of-mass motion [45]. For a dense ion ensemble, the frequency shift is observed due to a change in the local potential resulting from the space charge of the cloud [46]. Furthermore, the space charge from ion cloud lowers the effective confining potential, thereby reducing the amount of charge that can be stored in trap. If the cloud is assumed to be a nearly spherical ensemble of cold particles, the equations for frequency shift are given by [41]:

$$\nu_{z}' = \nu_{z} \sqrt{\left(1 - \frac{\nu_{p}^{2}}{3\nu_{z}^{2}}\right)} \quad \text{and} \quad \nu_{\pm}' = \frac{\nu_{c}}{2} \left(1 \pm \sqrt{\left(1 + \frac{2\nu_{p}^{2}}{3\nu_{z}^{2}}\right)\frac{2\nu_{z}^{2}}{\nu_{c}^{2}}}\right) \tag{2.23}$$

where ν_p is the plasma frequency given by $\nu_p^2 = q^2 n / \epsilon_0 m$, *n* is the number density of the cloud, ϵ_0 is the permittivity of free space and *m* is mass of the ion. The equation 2.23 gives the following conditions:

$$\frac{\nu_p^2}{\nu_z^2} < 3 \quad \text{and} \quad \frac{\nu_p^2}{\nu_z^2} < \frac{3}{2} \left(\frac{\nu_c^2}{2\nu_z^2} - 1 \right)$$
 (2.24)

which leads to the value for the maximum achievable particle density or the Brillouin limit as:

$$n_{max} = \frac{\epsilon_0 B^2}{2m} \tag{2.25}$$

The Brillouin limit is typically of the order of 10^9 charge per cm^3 for the magnetic field of a few Tesla. For a non-spherical cloud which fulfills the condition $2\nu_z^2 < \nu_c^2$, this can be achieved for any value of trapping potential. Further studies for different frequency shifts due to space charge effect can be seen in [43].

2.4 Ion manipulation and preparation for mass measurements

An advantage of the lack of rf-fields for the ion confinement in a Penning trap is that the rf-fields can be used for ion manipulation. Depending on the parameters of the rffields such as the applied frequency, amplitude, phase difference and excitation time, an ion can be manipulated in several ways. The external rf-fields for manipulation of radial motions are applied through segmented trap electrodes. The dipolar excitation manipulates individual ion motions while the quadrupolar excitation couples the eigenmotions causing conversion between the two modes of oscillation.

Dipolar excitation

Resonant electric dipolar excitation of the axial and radial modes of oscillation can be used to increase the amplitude of the respective oscillation. This can be used to determine the eigenfrequency of oscillation or for resonant ejection of unwanted particles from the trap. A dipolar field can be created by applying alternating potential at a given frequency at two symmetrically opposite electrodes. For the axial dipolar excitation, the rf voltage with frequency ν_z is applied to the endcap electrodes and for the radial dipolar excitation, the rf voltage at radial motional frequency (ν_+ or ν_-) is applied to the two opposite segments of the ring electrode.

A radial electric dipolar field created by applying an rf voltage with an amplitude U_d and a phase difference of π between two symmetrically opposite segments of a ring electrode will have x-component given by:

$$\vec{E_x} = \frac{U_d}{\rho_0} \cos\left(2\pi\nu_d t + \phi_d\right) \,\hat{x}$$
(2.26)

where ϕ_d is the initial phase of the dipolar excitation, ρ_0 is the radius of the ring electrode and \hat{x} is the unit vector in x-direction. The radial dipolar excitation at the segmented ring electrode with frequency $\nu_d = \nu_-$ excites all charged ions inside the trap to a larger radius, irrespective of their mass and charge due to mass independence of the magnetron motion (equation 2.11). However, dipolar excitation with $\nu_d = \nu_+$ can remove unwanted ion species by selective excitation of motional amplitudes due to the mass dependence of cyclotron frequency. If $\Delta \phi_- = \phi_d - \phi_-$ is the initial phase difference between the exciting dipolar field and the magnetron motion, then [47]:

- For $\Delta \phi_{-} = 0$, the radius increases slowly in the beginning and then almost linearly with time.
- For $\Delta \phi_{-} = 3\pi/2$, the amplitude of magnetron motion decreases initially and then increases linearly.
- For $\Delta \phi_{-} = \pi/2$, the magnetron motion amplitude follows a linear trend with t.

Figure 2.6 shows the variation of magnetron radius as a function of excitation time for different values of $\Delta \phi_{-}$.



Figure 2.6: Temporal evolution of calculated magnetron radius represented as a function of duration of dipolar excitation. This representation is for different values of phase difference between magnetron motion and dipolar excitation, $\Delta \phi = 0, \pi/2, 3\pi/2$. The figure is modified from [47].



Figure 2.7: Excitation geometries showing the segmentation of ring electrode for application of dipolar and quadrupolar excitation. In fig (a), the dipole field generation is shown by application of voltages with equal amplitude phase difference of half a cycle. In (b), the opposing segments have same voltages while the neighboring segments differ by 180 $^{\circ}$ in phase creating a quadrupolar field. The figure is modified from [47].

Quadrupolar excitation

A resonant electric quadrupolar excitation at a frequency equal to the sum or difference of individual eigenfrequencies of radial motion, couples the motions and enables interconversion between them. Such an excitation can be used to determine frequencies and for magnetron centering. In order to provide the quadrupolar excitation, a set of two opposing electrode segments (relative to trap center) are supplied with rf potential and the second pair of opposing electrodes are provided the same voltage amplitude but phase shifted by 180° relative to the first pair. In this case, the ring electrode is considered to have 4 segments with the neighboring electrode segments having a phase shift of half a cycle (as shown in figure 2.7). The quadrupole field is thus given by:

$$\vec{E_x} = \frac{2U_q}{\rho_0^2} \cos\left(2\pi\nu_q t + \phi_q\right) y \ \hat{x},$$
(2.27)

$$\vec{E_y} = \frac{2U_q}{\rho_0^2} \cos\left(2\pi\nu_q t + \phi_q\right) x \ \hat{y}$$
(2.28)

where U_q is the amplitude of quadrupolar excitation, ν_q is the excitation frequency, ϕ_q is the initial phase and \hat{x} and \hat{y} are the unit vectors in x and y directions, respectively. A full periodic conversion between the modified cyclotron and the magnetron motion (Rabi oscillations) takes place when quadrupolar excitation frequency is equal to the true cyclotron frequency [44].

$$\nu_q = \nu_c = \nu_+ + \nu_- \tag{2.29}$$

The conversion process between the two radial modes can be seen in figure 2.8. Let's consider a case where the charged particle is in pure magnetron motion. When



Figure 2.8: Interconversion of magnetron motion to a modified cyclotron motion at $\nu_q = \nu_c$. In figure (a), the ion motion is purely magnetron as shown by the circle around the center. Due to the quadrupolar excitation, the magnetron radius reduces and the cyclotron radius increases. Eventually after T_{conv} , the final cyclotron radius is equal to the initial magnetron radius. (a) and (b) show the first and second half of the conversion process. The figure is taken from [47].
the quadrupolar excitation is applied at $\nu_q = \nu_c$, the magnetron radius starts to decrease and the modified cyclotron radius increases. After a certain time $T_{conv} = 4\pi\rho_0^2 B/U_q$, the particle only undergoes modified cyclotron motion with its radius equal to the initial magnetron radius. Here, T_{conv} is the time needed to convert from one eigenmotion completely to the other. In the case of a non-resonant conversion, $\nu_q \neq \nu_c$, only a partial interconversion will occur [48].

2.5 Ion cooling in a Penning trap

The Penning trap is a widely used tool in the experiments aiming towards highprecision measurements. Cooling of the ion motion of the particles confined in a Penning trap reduces the Doppler broadening and facilitates longer storage times, easier ion cloud manipulations and high-precision measurements [49]. The term 'cooling' used here refers to the reduction of particle energies and hence their oscillation amplitudes. Many cooling techniques have been developed and utilised for Penning trap experiments such as resistive cooling [50], laser cooling [49], buffer gas cooling [51] and evaporative cooling [52]. A brief overview of the cooling techniques used in the high-precision Penning trap experiments ARTEMIS and SHIPTRAP is presented in this section.

2.5.1 Buffer gas cooling

If a neutral buffer gas is present along with the charged particles, the collisions between them lead to equilibration of the particle energy which can be used for particle cooling. This cooling technique is known as buffer gas cooling. An advantage of this technique is its applicability over a broad range of energies and oscillation frequencies. When a particle of mass m and charge q is exposed to a residual buffer gas in a Penning trap, it experiences a damping force proportional to its velocity v, given by:

$$F = -\delta mv \quad \text{with} \quad \delta = \frac{q}{m} \frac{1}{M_0} \frac{pT_0}{p_0 T}$$
(2.30)

where δ is the damping coefficient dependent on M_0 , which is the particle's reduced mobility in a buffer gas at $p_0 = 1013$ mbar and $T_0 = 300$ K [53, 54]. The damping of the radial motions due to the reduction of axial particle motion is characterised by the initial radii $\rho_{\pm}(0)$, which evolve with time as:

$$\rho_{\pm}(t) = \rho_{\pm}(0) \exp\left(\mp \frac{\nu_{\pm}}{\nu_{+} - \nu_{-}} \delta t\right)$$
(2.31)



Figure 2.9: An illustration of the radial ion motion during buffer gas cooling in a Penning trap. (a) In the absence of quadrupolar excitation, the cyclotron radius reduces faster than the increase in magnetron radius. (b) By coupling both radial modes using a quadrupolar excitation at $\nu_q = \nu_c$, the cyclotron and the magnetron radii decrease and the ion is centered in Penning trap. The figure is modified from [55].

Since the buffer gas cooling reduces the energy of all three motions at the same time, the amplitude of axial and reduced cyclotron motion decreases, but the magnetron motional amplitude increases. This can lead to particle loss through an increase in the magnetron radius. As $\nu_+ \gg \nu_-$, the decrease in modified cyclotron radius is much faster than the increase in magnetron radius. The ion loss due to constantly increasing magnetron radius can be avoided by resonant coupling (as discussed in the framework of magnetron centering, section 5.5). At the SHIPTRAP set-up, this ion loss is avoided by applying a quadrupolar excitation at free cyclotron frequency ν_c of the ion of interest (section 2.4) which effectively leads to a centering of ion motion in the Penning trap (figure 2.9). The free cyclotron frequency depends on the q/m value, thus making the buffer gas cooling a mass-selective process. In the preparation trap of SHIPTRAP (chapter 6), the buffer gas technique is used allowing isobaric purification of the ion of interest with a mass resolving power ($\nu_c/\delta\nu_c$) of the order of 10⁵ [56, 57].

2.5.2 Evaporative cooling

The kinetic energy of a particle is a measure of its temperature. Trapping of particles with higher kinetic energy requires deeper potential wells. When the trap depth is lowered, ions with highest kinetic energy are kicked out of the trap and the remaining trapped particles equilibrate their energy to a lower value. Therefore, the ions are



Figure 2.10: An illustration of the principle of evaporative cooling. When the trap depth is reduced, the hotter particles leave the trap, thereby leading to a lower equilibrium temperature of the remaining ensemble.

thus cooled. This technique is known as 'evaporative cooling'. This is a common technique in traps for neutral particles and is also applied for the cooling of highly charged ions in electron-beam ion traps (EBITs) and in Penning traps. The technique has an advantage of being applicable on any charged particle and is easy to implement. However, a side-effect of this cooling approach is that the particles are lost during the process and the final number of particles is reduced. The thermal distribution of an ion ensemble can be considered to have a Maxwell-Boltzmann distribution, with the probability function given by:

$$p(E)dE = \sqrt{\frac{4E}{\pi}} \left(\frac{1}{k_B T}\right)^{3/2} \exp\left(-\frac{E}{k_B T}\right) dE, \qquad (2.32)$$

where $\int_0^\infty p(E)dE = 1$ and a fraction of particles f with kinetic energies above a certain value E' is given by:

$$f = \int_{E'}^{\infty} \sqrt{\frac{4E}{\pi}} \left(\frac{1}{k_B T}\right)^{3/2} \exp\left(-\frac{E}{k_B T}\right) dE.$$
 (2.33)

Thus, after lowering the trapping potential to U = E'/q, a fraction f of the trapped particles leave the trap. Figure 2.10 illustrates the principle of evaporative cooling, with the hotter particles leaving the trap when the trap depth is lowered, thereby leading to a lower equilibrium temperature of the remaining ensemble.

2.5.3 Resistive cooling and detection

An oscillating charged particle in an ideal Penning trap can be considered as a harmonic oscillator. The motion of a particle confined in a Penning trap induces oscillating image currents on the confining electrodes. These induced currents are typically of the order of few fA to pA. In an ideal Penning trap, the ion motion can be described in terms of a series LC circuit [50]. This equivalent *lc* circuit connected to a voltage of U_p , has the capacitance and inductance of c_p and l_p , respectively, such that:

$$l_p \frac{dI_p}{dt} + \frac{1}{c_p} \int I_p dt = U_p$$
(2.34)

where I_p is the image current induced at the trap electrodes by this oscillating charged particle. For a cloud with N particles of a single ion species with charge q at a given coordinate ρ_i , this induced current [50, 58] is defined as:

$$I_p = \dot{\rho}_i \frac{Nq}{D} = 2\pi\nu_i \rho_i \frac{Nq}{D} \qquad ; i \in [z, +]$$

$$(2.35)$$

where ν_i is the oscillation frequency of the motion (ν_z, ν_+) and D is the effective electrode distance of the specific particle motion containing all the information about the geometry of the electrodes [35]. For an induced surface charge density $Q_i(\rho, z) =$ $-q \cdot \Xi(\rho, z)$ on the trapping electrode by an ion with charge q, the value of D in terms of the geometry function $\Xi(\rho, z)$ is given as:

$$D^{-1}(\rho, z) = \frac{\partial}{\partial z} \Xi(\rho, z)$$
(2.36)

with

$$\Xi(\rho, z) = \frac{-1}{\rho_0 \pi} \int_0^\infty \frac{\mathrm{I}_0 (x\rho/\rho_0)}{\mathrm{I}_0 (x)} \left[\operatorname{sinc}(x \frac{z - z_F}{\pi \rho_0})(z - z_F) - \operatorname{sinc}(x \frac{z - z_N}{\pi \rho_0})(z - z_N) \right] dx.$$
(2.37)

Here, $I_0(x)$ is the modified Bessel function of the first kind, z_N and z_F are the distance of the center of the trap to the nearest and the farthest edge of the pick-up electrode and ρ_0 is the inner radius of the cylindrical trapping electrodes.

In order to detect the small induced currents (equation 2.35), an inductor L_p is connected to the electrodes. This coil with inductance L_p has a self capacitance C_p and a resistance R_p and hence forms a parallel RLC circuit or a 'Resonator'. The self capacitance of this resonator is mainly due to capacitance between the coil turns and also from the capacitance between the coil and the resonator housing. This depends on various factors such as geometry of the resonator, number of turns, insulation material of the wire and the material used to produce the resonator housing [58, 59] (sections 4.4). A schematic of the parallel RLC circuit connected to one of the trap electrodes is shown in figure 2.11.

When the motional frequency of an oscillating ion confined in the Penning trap is the same as the eigenfrequency of the resonator (i.e. the ion is in resonance with the RLC circuit), the impedance of the circuit is maximum with $R_p = 2\pi\nu_0 LQ$. Therefore, power $P = I_p^2 R_p$ is transferred into the resistance of the resonance circuit (R_p) via the image current (I_p) [60]. The RLC circuit, which is kept at liquid helium temperature, acts as a heat sink and the ion motion can be cooled to that temperature. This technique of cooling by dissipation of energy into an RLC circuit is known as 'resistive cooling'. The decay in kinetic energy for a single ion is exponential (assuming frictional loss model) and for the axial motion can be expressed as:

$$E_z = E_z(0) \exp(-\gamma t), \quad \text{with} \quad \gamma = \frac{q^2}{m} \frac{R_p}{D^2}$$
(2.38)

where γ is the energy damping constant and is related to cooling time constant as $\tau_z = \gamma^{-1}$. For an ensemble with N particles, each having a charge q and a mass m and oscillating in the same phase, the cooling time constant is given by [41]:

$$\tau_N = \frac{1}{N} \frac{m}{R_P} \frac{D^2}{q^2} = \frac{1}{N} \tau_z.$$
 (2.39)

This indicates that such an ion cloud cools N times faster than a single ion. However, for an ensemble with N particles having arbitrary phases, the time average of the mean square value for the induced current is taken into account. This gives the cooling time constant which is same as the value for a single ion. Therefore, the cooling of an ensemble of N ions with random phases is the same as cooling an ion with charge Nq and mass Nm or cooling the center-of-charge of that cloud [61].

Resistive cooling and non-destructive detection of a particle's oscillatory motion are two different aspects of the same process of dissipation of energy through an RLC circuit. The dissipation of energy by the induced image current through the resistive circuit cools the particle motion, representing resistive cooling. Concurrently, the image current through the resonant circuit produces a detectable time-dependent voltage signal which can be analysed using Fourier transform to obtain the frequency spectrum. A parallel RLC circuit is commonly used for resonant cooling and nondestruction detection of the ions confined in the Penning trap (schematic in figure 2.11).

The frequency of resonance of the RLC circuit with inductance L_p and self capacitance C_p is given by:

$$\nu_0 = \frac{1}{2\pi\sqrt{L_p C_p}}.$$
(2.40)

Apart from the self-capacitance, the trap electrodes and connecting wires also contribute to the total capacitance of the system. These are termed as trap load capacitance C_T . This acts as a parallel load capacitance which shifts the central frequency of the resonator to a value:

$$\nu_0 = \frac{1}{2\pi\sqrt{L_p(C_p + C_T)}}.$$
(2.41)



Figure 2.11: Schematic of a typical non-destructive ion detection and cooling system. A parallel RLC circuit is connected to one of the electrodes of a cylindrical Penning trap followed by the amplifiers. R_p , L_p and C_p are the effective resistance, inductance and self capacitance of the coil. When the ion motion frequency is the same as the resonance frequency of the circuit, a voltage drop occurs, which is further amplified by the cryogenic and room temperature amplifiers. This amplified signal is displayed by the spectrum analyser.

For any RLC circuit, the quality factor, also known as 'Q-factor', is an important parameter that needs to be taken care of while designing a resonator. The Q-factor is a measure of efficiency of electrical components and is defined as the ratio between the stored energy in a resonator and the dissipated energy per oscillation cycle. It can also be described as the ratio of resonance frequency ν_0 and the spectral width $\Delta\nu$ of the resonator:

$$Q = \frac{\nu_0}{\Delta\nu}.\tag{2.42}$$

When the resonance spectrum from the RLC circuit is observed at the spectrum analyser, ν_0 corresponds to the central frequency and $\Delta\nu$ is the full width at half maximum (FWHM) of the Lorentzian. From the spectrum observed, $\Delta\nu$ can be measured at -3 dB m from the amplitude of resonance frequency (figure 2.12).

At the resonance frequency ν_0 , the resonator has an effective parallel impedance given by:

$$R_p = 2\pi\nu_0 LQ \tag{2.43}$$

Therefore, at the resonant frequency, when the frequency of the RLC circuit



Figure 2.12: Lorentz spectrum response for a resonator circuit showing the central frequency ν_0 and the peak width at -3 dB m relative to the peak amplitude at resonance frequency ν_0 .

matches the motional frequency of the charged particle, the observed voltage drop is

$$U_p = R_p I_p = 2\pi \nu_0 L_p Q I_p.$$
 (2.44)

As depicted in equation 2.43, it is preferable to have resonators with high Q-factor and inductance values in order to maximize the voltage drop U_p . After amplification from cryogenic amplifiers (section 4.4.2) and room-temperature amplifiers [43], the voltage is Fourier transformed to obtain the frequency spectrum of the particle at the spectrum analyser. For the Fourier transformed signal in the frequency space, a dip is observed at the central frequency of the resonator if the motional temperature of the ion is the same as the temperature of the LC-circuit. However, for a hot ion, a peak is created on top of the resonator signal at the ion oscillation frequency. At ARTEMIS, the measurements are performed in the voltage space by taking q/m scans as described in section 5.3.

The final quality factor of the detection system depends on the Q-factor of each of the components comprising the system. As can be seen in equation 2.45, the overall quality of the system, Q_f , is limited by the component with lowest Q-factor [62].

$$\frac{1}{Q_f} = \frac{1}{Q_{\text{Res}}} + \frac{1}{Q_{\text{Amp}}} + \frac{1}{Q_{\text{Trap}}} + \frac{1}{Q_{\text{Var}}} + \frac{1}{Q_{\text{Con}}} + \dots$$
(2.45)

Here, Q_i , $i \in [Res, Amp, Trap, Var, Con]$ corresponds to the quality factor of the resonator, amplifier boards, trap's pick-up electrodes, varactor diode boards and connectors (feedthroughs, connection cables), respectively. The components listed here comprise the non-destructive detection system of the Penning trap experiments such as ARTEMIS. The varactor diodes are used in the detection system to provide flexibility in the central frequency of the resonator.

Equation 2.45 further strengthens the need for a high quality resonator coil. The resonator should be designed to resonate at a particular resonance frequency given

by the details of the experiment. There are various factors that should be taken care of when designing a resonator for the non-destructive detection technique in the Penning traps, such as the space considerations, the strong magnetic fields and the cryogenic temperature. There are two main designs of inductor coils used in trapping experiments, i.e. helical and toroidal coils. The resonators are commonly fabricated by winding a normal-conducting or superconducting wire around an insulator (usually made from Polytetrafluoroethylene (PTFE) or teflon) and placing it in a highly conductive and fully closed housing (usually made from oxygen-free highconductivity copper (OFHC), sometimes gold-plated). The details on the resonators used at ARTEMIS within the scope of this work are discussed in section 4.4.

In contrast to the non-destructive detection of ion motion in the ARTEMIS Penning trap using an RLC circuit, the micro-channel-plate (MCP), channeltron and time-of-flight (ToF) detectors are implemented in the SHIPTRAP set-up to destructively detect and record the position and timing information of the confined ions. The details of these detectors are given in section 6.5.

Chapter 3

Theory and principle of measurement

The measurement of intrinsic properties such as mass and magnetic moment of the elementary particles, is an effective method to probe the nuclear structure and properties, and test the Standard Model of particle physics. The measurements of magnetic moments with high-precision has led to the studies of anti-matter [63, 64] and to the significant improvement in the fundamental constants such as α , the fine-structure constant [7, 8, 65]. Precise mass measurements of radioactive nuclides help to reveal information on their nuclear structure [19, 66]. A precise tool to obtain these properties in elementary particles is the Penning trap. This chapter describes the theoretical motivation and the principle of measurement for two Penning trap experimental setups at GSI, Germany: the ARTEMIS experiment that aims to measure the magnetic moment of a bound electron in heavy, highly charged ions, and the SHIPTRAP experiment for direct mass measurements of heavy and superheavy nuclides.

3.1 g-factor of the free electron

The magnetic moment of a free electron with the intrinsic spin angular momentum \vec{s} , can be written as:

$$\vec{\mu}_s = -g_s \mu_B \frac{\vec{s}}{\hbar} \tag{3.1}$$

where the g-factor, g_s , is a dimensionless constant which connects the magnetic moment $\vec{\mu_s}$ to its angular momentum in terms of the reduced Planck's constant $\hbar = h/2\pi$ and the Bohr magneton μ_B defined by:

$$\mu_B = \frac{e\hbar}{2m_e} \tag{3.2}$$



Figure 3.1: Feynman diagram for the first-order QED contribution to the g-factor of a free electron. This is also referred to as the Schwinger term. The diagram represents the self-energy term with the emission and absorption of a virtual photon. The black triangle represents the interaction with an external magnetic field.

where e and m_e are, respectively, the charge and mass of the electron. The g-factor for a free electron was predicted within Dirac theory to be exactly equal to 2. However, deviation from this value has been observed, giving:

$$\frac{g_s}{2} = 1 + a_{\text{QED}} + a_{\text{hardonic}} + a_{\text{weak}} \tag{3.3}$$

where the first term comes from the Dirac equation. The contributions due to the interaction of an electron with the hadron-antihadron pairs and with the weak bosons are predicted to be smaller than 2×10^{-12} [41]. The deviations are dominated by the QED contribution (0.1%), arising due to the interaction of the free electron with the electromagnetic field. Based on the Feynman diagrams, the QED contribution can be calculated by an expansion in terms of the fine-structure constant $\alpha \approx 1/137$, such that:

$$a_{\text{QED}} = C_2 \left(\frac{\alpha}{\pi}\right) + C_4 \left(\frac{\alpha}{\pi}\right)^2 + C_6 \left(\frac{\alpha}{\pi}\right)^3 + \dots$$
(3.4)

Here, the expansion terms are based on the order of the Feynman diagram. As an example, the first term which is linear in α corresponds to the first-order Feynman diagram (having 2 vertices) with the corresponding coefficient C_2 calculated by J. Schwinger to be 0.5 [67]. The corresponding first-order diagram with selfenergy energy term has been shown in figure 3.1. In the same way, for the calculation of higher order coefficients, all the possible corresponding Feynman diagrams need to be computed. The number of these possible combinations rises rapidly, with C_4, C_6, C_8 and C_{10} requiring 7, 72, 891 and 12672 Feynman diagrams to be computed, respectively.

3.2 The case of the bound electron

The interactions of the electron bound in an atomic system are more complex in comparison to the free electron. Along with the terms described in equation 3.3, some other effects have to be taken into account such that [41]:

$$g = g_D + \Delta g_{\text{QED}} + \Delta g_{\text{int}} + \Delta g_{\text{SQED}} + \Delta g_{\text{nucl}}$$
(3.5)

where g_D is the leading order Dirac-Breit term [68], Δg_{QED} is the one-electron QED contributions, Δg_{int} is the interelectronic-interaction contribution, Δg_{SQED} is the screened QED contribution and Δg_{nucl} is the nuclear contribution. These are discussed here briefly.

- g_D is the Dirac term for a bound electron interacting with the nuclear Coulomb potential.
- Δg_{QED} is parallel to the QED contributions for a free electron, which arises from the Feynman diagrams of self-energy, vertex corrections and vacuum polarisation. The diagram for the bound electron is represented by a double solid line indicating that the electron always propagates in the Coulomb field of the nucleus. The expansion is made in αZ and the number of Feynman diagrams for each order is much higher than the free electron case. For the first-order, there are six diagrams that need to be computed in contrast to only one for the free electron. However, for the second order this number increases rapidly to 50 bound electron Feynman diagrams. The first-order one-electron QED diagrams are shown in figure 3.2.
- Δg_{int} accounts for the contribution due to exchange of photons between the electrons. The contribution is of the order of $(\alpha Z)^2/Z^n$, where *n* is the number of exchanged photons. Many-electron photon exchange has to be taken into account for the systems with more than 2 electrons.
- Δg_{SQED} corresponds to the contribution due to the Feynman diagrams with vacuum polarisation and self-energy interactions for multi-electron systems. To bypass the rigorous calculations for the two electron QED diagrams, one-electron QED contributions with a screened potential can be computed [41].
- Δg_{nucl} is the sum of the contributions due to finite nuclear size, nuclear recoil and nuclear polarisation. The distribution of the nuclear charge leads to a modification of the nuclear potential, thereby introducing finite size effects. These can only be calculated with uncertainties given by the knowledge of the nuclear size [9]. In the Furry picture of QED, finite nuclear mass was not considered. However, the nuclear recoil effects result due to the consideration of finite mass of the nucleus. Nuclear polarisation contributions arise from the nuclear excitations due to electromagnetic coupling to the electrons. These polarisation contributions are generally small and have been investigated in detail in [69].



Figure 3.2: (a) The first-order Feynman diagrams for the bound electron. The diagram represent the self-energy and vacuum polarisation terms. In these Feynman diagrams, a double-line represents a bound electron propagating in the Coulomb field of the nucleus. (b) Expansion in αZ of the different interaction terms such as self-energy, vacuum polarisation and vertex correction.

g-factor of the bound electron

For an electron bound in an atomic system, the orbital angular momentum \vec{l} and the spin \vec{s} of the electron couple together to give the total angular momentum $\vec{j} = \vec{l} + \vec{s}$. Due to the spin-orbit coupling, the individual direct determination of g_l and g_s are not possible. The g-factor corresponding to the total angular momentum (g_j) can be described in terms of g_l and g_s as:

$$g_j = g_l \frac{j(j+1) - s(s+1) + l(l+1)}{2j(j+1)} + g_s \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)}$$
(3.6)

Similar to equation 3.1, the g-factor (g_j) of a bound electron relates the magnetic moment $\bar{\mu}_j$ to the total angular momentum \vec{j} as:

$$\vec{\mu}_j = -g_j \mu_B \frac{\vec{j}}{\hbar} \tag{3.7}$$

The presence of an external magnetic field $\vec{B} = B_0 \hat{z}$, lifts the degeneracy in the magnetic quantum number of the fine-structure states and splits it into 2j+1 Zeeman sub-states. The energy difference between two consecutive Zeeman sub-states is given by:

$$\Delta E = h\nu_L = g_j \mu_B B_0 \tag{3.8}$$

where ν_L is the Larmor frequency of precession of the electron spin in the external magnetic field. Therefore, in order to have a high-precision measurement of g_j , accurate knowledge of ν_L and B_0 is required. In addition to the Larmor precession of the



Figure 3.3: Schematic of a bound electron in a heavy, highly charged hydrogen-like ion. The electron precesses around the magnetic field axis with the Larmor frequency ν_L and the ion moves in a circular motion about the field axis with the cyclotron frequency ν_C (ν_C << ν_L). The extreme electromagnetic field of the nucleus modifies the *g*- factor of the bound electron relative to the free electron.

electron, the ion also undergoes circular motion about the magnetic field lines with the cyclotron frequency ν_c , given by:

$$\nu_c = \frac{qB_0}{2\pi m} \tag{3.9}$$

where q and m are, respectively, the charge and mass of the ion. Since, frequency is one of the parameters that can be measured precisely, hence the value of B_0 can be obtained in terms of the cyclotron frequency of ion motion using equation 3.9.

Therefore, the g-factor for a bound electron can be represented in terms of the ratio of Larmor and cyclotron frequency as:

$$g_j = 2\frac{\nu_L}{\nu_c} \frac{q}{e} \frac{m_e}{m} \tag{3.10}$$

Figure 3.3 shows the schematic of a heavy, highly charged hydrogen-like ion in an external magnetic field. The bound electron is present in the extreme fields of the nucleus which changes its μ_j . In ARTEMIS, the Larmor frequency can be measured by using the double-resonance spectroscopy technique (section 3.3) and the cyclotron frequency measurements are performed using the non-destructive detection technique and sideband coupling (section 2.5.3).

3.3 Laser-microwave double-resonance spectroscopy

In the conventional precision experiments for the g-factor measurement, the Larmor frequency is measured using a continuous Stern-Gerlach scheme [70]. In contrast to this, ARTEMIS utilises the laser-microwave double-resonance spectroscopy technique. Although, the double-resonance technique has already been applied for measuring the g-factor of the atoms and singly charged ions [71, 72], it will be used for the first time at ARTEMIS for the measurement of magnetic moments in highly charged ions. There are two significant advantages of the use of double-resonance technique: a) it allows for the measurement of the nuclear magnetic moment and the bound electron magnetic moment simultaneously in one Penning trap and b) the absence of an electron cloud in the highly charged ions facilitates the measurement of nuclear magnetic moments in the absence of diamagnetic shielding effects.

In the singly charged ions, the fine-structure and hyperfine-structure transitions are in the microwave domain. On the contrary, for the highly charged ions, strong electric and magnetic fields of the nucleus shifts these transitions into the optical and ultraviolet regime. The energy of the fine-structure transitions scales as Z^4 , thereby making the ions in the medium Z range as potential candidates for transitions in the laser accessible region. Similarly, for the hyperfine-structure splitting, ions in the range of high Z are suitable candidates since the transition energy scales as Z^3 [73].

As the first candidate for the application of laser-microwave double-resonance spectroscopy technique, boron-like argon (Ar¹³⁺) is chosen. The ion has no nuclear spin and a completely filled 1S and 2S shells with one valence electron in the 2P shell. Due to the spin-orbit coupling, the 2P electron has fine-structure doublet states $2^2P_{1/2}$ and $2^2P_{3/2}$. In an external magnetic field, the m_j degeneracy is lifted and the two fine-structure states split into six Zeeman sub-states as shown in the figure 3.4. The fine-structure doublet is separated by an energy of ~2.8 eV corresponding to a laser accessible wavelength of 441.25575(17) nm [74] with a lifetime of 9.573(6) ms [75] for the excited fine-structure state $2^2P_{3/2}$. The transitions between the Zeeman sub-states are in the microwave domain with 130 GHz and 65 GHz, respectively, for $2^2P_{3/2}$ and $2^2P_{1/2}$ sub-states (figure 3.4).

When a closed optical cycle between $|1/2, +1/2\rangle$ and $|3/2, +3/2\rangle$ is probed by an optical laser with frequency ν_{op} , roughly half of the electrons from $|1/2, +1/2\rangle$ populate the state $|3/2, +3/2\rangle$. The electrons in the excited state $|3/2, +3/2\rangle$ undergo spontaneous decay to the lower state $|1/2, +1/2\rangle$ through a magnetic dipole (M1) transition. Thus, a fluorescence signal is observed which can be detected using a camera (section 4.6). Subsequently, a tunable microwave signal around ν_{mw1} is shone into the trap while monitoring the fluorescence signal. When the frequency of the microwave radiation resonates with the Larmor frequency ν_L , the electrons undergo a stimulated transition to the state $|3/2, +1/2\rangle$. The electrons in this state can decay through two possible channels to the states $|1/2, -1/2\rangle$ and $|1/2, +1/2\rangle$. This results in a decrease in the observed fluorescence light due to the population of the dark state $|1/2, -1/2\rangle$. Thus, the microwave frequency at which the fluorescence intensity



Figure 3.4: Level structure foreseen for the laser-microwave double-resonance spectroscopy of (a) 40 Ar¹³⁺ (b) 209 Bi⁸²⁺. In figure (a) ν_{op} is the fine structure closed-transition accessible in the optical region (corresponding to 441 nm) for 40 Ar¹³⁺. The microwave radiation with frequency $\nu_{mw1} \approx 130$ GHz causes the transition to a dark state since it can decay via the two modes, shown by the red lines. For figure (b) the hyperfine structure transitions in 209 Bi⁸²⁺ is represented by ν_{UV} and the Zeeman transitions by ν_{mw1} and ν_{mw2} .

reduces gives the Larmor frequency of precession of the electron bound to Ar^{13+} . Figure 3.4 shows the Zeeman splitting and highlights the transition involved in the double-resonance technique for ${}^{40}Ar^{13+}$ and ${}^{209}Bi^{82+}$.

²⁰⁹Bi⁸²⁺ is an excellent species for double-resonance spectroscopy. Contrary to argon, ²⁰⁹Bi has a nuclear spin of I = 9/2, thereby leading to hyperfine-structure splitting with transitions at 243.87(2) nm [76] and lifetime of 399.5(1.8) µs [77]. Again, the presence of magnetic field lifts the m_F degeneracy, and the hyperfine state with total angular momentum F splits into 2F+1 Zeeman sub-states separated by ~20 GHz [12]. In the same way as the boron-like argon, the double-resonance spectroscopy can be implemented on ²⁰⁹Bi⁸²⁺ by probing the closed transition between $|4, -4\rangle$ and $|5, -5\rangle$ states with a laser. The optical signal from the closed transition is observed while scanning the microwave frequencies ν_{mw1} and ν_{mw2} . A decrease in the optical signal indicates the reduction in the population of the closed cycle, thereby determining the Larmor frequency.

Thus, laser-microwave double-resonance spectroscopy technique can be used in order to achieve high-precision g-factor measurements and provide the most stringent test of QED in strong fields.

3.4 Higher-order Zeeman effect

The energy states of an ion undergo Zeeman splitting when subjected to external magnetic fields. In the presence of high magnetic field (7 T at ARTEMIS), the nonlinear



Figure 3.5: Higher-order Zeeman level scheme for boron-like argon, Ar^{13+} .

higher-order splittings also become significant. The Zeeman split energy sub-states are equidistant in the first-order approximation but the non-linear magnetic-field effects perturb this symmetry for higher orders. These perturbed energy states are given by:

$$E_A(B_0) = E_A^{(0)}(B_0) + \Delta E_A^{(1)}(B_0) + \Delta E_A^{(2)}(B_0) + \Delta E_A^{(3)}(B_0) + \dots$$
(3.11)

where $|A\rangle = |j, m_j\rangle$ is the $2^2 P_j$ state with angular momentum j and its projection m_j . $E_A^{(0)}$ is the energy in the absence of external magnetic field. The first-order shift corresponds to the anomalous Zeeman effect with equidistant splitting and can be related to g_J as:

$$\Delta E_A^{(1)}(B_0) = g_J \mu_B B_0. \tag{3.12}$$

 Ar^{13+} is a good candidate to study these effects in highly charged ions due to the profound theoretical knowledge of its energy levels splittings [68]. Figure 3.5 shows the higher-order Zeeman level scheme for boron-like argon, Ar^{13+} . The second-order shifts are of the order of kHz or MHz, which are within the expected resolution of measurement. Hence, for the high-precision g_j measurement for Ar^{13+} , these contributions should be disentangled by measuring various optical and Zeeman states. Techniques to measure and disentangle the higher-order shifts from the desired frequency have been described in detail in [34].

3.5 Motivation for mass measurements at SHIPTRAP

The atomic nucleus consists of protons and neutrons. Inside the nucleus there are two competing forces acting on the nucleons, the strengths of which determine the stability of the nucleus: a) the strong nuclear forces which tend to hold the nucleons together and b) the repulsive Coulomb force among the protons which tends to disintegrate the nucleus. The attractive strong forces are short-range and charge-independent in contrast to the repulsive Coulomb forces that act between the charged particles and are long range. The stability of the nucleus is determined by its binding energy, which is the energy required to break the nucleus into its constituents. For a nucleus with mass number A = Z + N, the binding energy can be quantified as the mass defect, i.e the difference between the experimentally obtained mass and the mass of its constituents.

There are several models that predict nuclear properties including the binding energy. The liquid drop model is based on the approximation that each nucleon interacts with its nearest neighbor, as in a drop of an incompressible homogeneous liquid, kept together by the nuclear force [78]. While it reproduces the general trend of the binding energy for most nuclei, it does not explain the increased stability for the so-called 'magic numbers'. The occurrence of these magic nuclei is explained by the shell model [79, 80], according to which only discrete energy levels can be occupied by the nucleons. The model is based on the assumption that a nucleon moves independently in a three-dimensional nuclear potential formed by all the other nucleons. The form of this potential is not precisely known, hence the Woods-Saxon potential [81], based on the nucleon density distribution, is often used for calculations. The shell model explains the occurrence of magic nuclei and the existence of superheavy nuclei ($Z \ge 104$). Unfortunately, none of the nuclear models provides a complete description of the nuclear structure and properties. In addition to this, different theoretical models predict different locations for the 'island of stability' in the region of superheavy elements with the possible shell closures at N = 172 or 184 and Z = 114, 120 or 126 [82, 83, 84]. The 'island of stability' refers to a predicted set of isotopes of the superheavy elements with considerably longer half-lives than the known isotopes of these elements.

Accurate knowledge of the masses of the superheavy elements is an important factor that helps to constrain some of the parameters in theoretical models and to explore the nuclear structure at the boundary of nuclear existence. The binding energy of a nucleus is a model-independent quantity and provides insight into its structure. From the direct high-precision mass measurements, the binding energy of nuclei can be directly obtained.

The SHIPTRAP mass spectrometer located at the GSI facility in Darmstadt, Germany, is a Penning trap experiment that enables high-precision measurements of superheavy and exotic nuclei with rather short half-lives of about 200 ms and above. There have been various mass measurements of exotic heavy nuclei at the SHIPTRAP set-up [85, 86] with the most recent ones comprising 251,254 No, 254,255,256 Lr and 257 Rf, with a relative precision of $\delta m/m \sim 10^{-9}$ [87]. The high mass resolving power of the phase-imaging ion-cyclotron-resonance technique ($\sim 10^7$) also allows resolving nuclear isomers with low excitation energy (about 40 keV for A = 250).

Due to the low production rates and short half-lives, performing direct highprecision measurements on even heavier elements is not trivial. However, the study of lighter nuclides aids in benchmarking the nuclear model to make predictions of properties for heavier nuclei.

An extensive web which links the masses of all the known nuclides and provides a comprehensive set of masses has been given in the form of the Atomic Mass Evaluation. The masses of the nuclides are either directly measured or are 'indirectly' derived from the studies of radioactive decays or nuclear reactions. In order to achieve accurate masses from the decay chains up to the heaviest elements, more experimental data is needed by direct mass measurements. The data is regularly updated, with the most recent publication in this series being the AME 2020 atomic mass evaluation [88]. As discussed in the recent publication by O. Kaleja *et al.* [87], the high-precision mass measurements of ^{251,254}No, ^{254,255,256}Lr and ²⁵⁷Rf serve as new anchor points and improve 15 masses up to ²⁷⁰Ds.

For these direct-mass measurements, the efficiency of the cryogenic gas cell (CGC, section 6.2) of the SHIPTRAP experiment plays an important role. Recoil-ion sources are installed in the CGC to characterise and optimise the gas-stopping cell [89]. Considering the requirements and characteristics for a recoil-ion source (section 7.1), the choice of species leads to 225 Ac and 223 Ra. This comes with an unexpected benefit: since there are not many direct mass measurements in a vast region of the nuclear chart¹, high-precision direct mass measurements in this region with Z = 80 to Z = 90 are of great interest. The 205 Tl / 205 Pb doublet masses are linked to 207 Tl and 207 Pb through nuclear reactions. These 207 Tl and 207 Pb are the daughter nuclides from the recoil-ion sources installed in cryogenic gas cell (figure 7.1). Therefore, the measurements on the linked elements are crucial for the Q-value measurement of this doublet

¹The masses are mostly determined by reactions and decays.

which finds its applications in nuclear astrophysics for the calculation of nuclear matrix elements and as s-process cosmochronometer [22]. Furthermore, the masses of the decay products from recoil-ion sources are linked to the mass of 209 Bi, which is the prime candidate for a high-precision g-factor measurement in the ARTEMIS experiment at GSI, Germany. The uncertainty in the mass of 209 Bi contributes directly to the total error budget of the g-factor measurements (section 3.2).

In the present work, mass measurements are performed for different nuclides obtained in the decay chains of the recoil-ion sources ²²⁵Ac and ²²³Ra installed in the cryogenic gas cell of SHIPTRAP in an attempt to improve their present mass uncertainty. The phase-imaging ion-cyclotron resonance technique (PI-ICR, section 3.6) has been used to determine the masses of ²²¹Fr, ²¹³Bi and ²⁰⁹Pb from the decay chain of ²²⁵Ac and ²¹⁹Rn, ²¹¹Pb, ²⁰⁷Tl and ²⁰⁷Pb from the decay chain of ²²³Ra. These mass measurements establish new anchor points in the decay chains and provide input data to improve the theoretical predictions regarding the properties of heavier elements [90]. Due to the high-precision mass measurements, these elements also provide new references for other direct mass measurements.

3.6 Phase-Imaging Ion-Cyclotron-Resonance technique

In order to measure the cyclotron frequency of an ion confined in a Penning trap, the Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique [20] has been developed at the SHIPTRAP experiment at GSI. In comparison to the conventional time-of-flight ion-cyclotron resonance (ToF-ICR) technique [48] under similar experimental conditions, the mass resolving power is increased by up to a factor of 40 and the precision by up to a factor of 5. As explained in the subsequent sections, lesser number of ions are needed for obtaining a particular level of statistical precision using the PI-ICR technique. Hence, it is about 25 times faster as compared to the ToF-ICR method. The technique is based on the projection of the phases of the radial modes of an ion (cyclotron and magnetron motion) onto a position-sensitive detector for two different ion confinement times in the Penning trap.

Based on the excitation pattern used in the PI-ICR technique, it can be classified into two categories: single-pattern scheme for independent measurement of the radial frequencies ν_+ and ν_- and double-pattern for direct determination of the cyclotron frequency [20]. Figure 3.6: Projection of the radial motion of the ions in a Penning trap onto a position-sensitive detector. The center image is obtained without any excitation (figure 3.7). The final phase image corresponds to radially excited ions after accumulating a phase during time t_{acc} , while the reference phase image is obtained by applying the extraction pulse shortly after radial excitation [91].



3.6.1 Single-pattern scheme: independent measurements of ν_{-} and ν_{+}

Using the single-pattern PI-ICR scheme, the free cyclotron frequency of an ion can be determined by measuring the individual radial motional frequencies, since they follow the relation $\nu_+ + \nu_- \approx \nu_c$ [38]. A cooled ion without radial energies is then injected into the trap. For the magnetron frequency measurement (as can be seen in figure 3.7), the ion is radially excited to a well-defined radius r_- when subjected to a dipolar excitation with frequency ν_- and amplitude A_- . The ion is now in a pure magnetron motion with frequency ν_- and traces a circular orbit with radius r_- . If an ejection pulse is applied at time t_0 , the detected ion spot acts as the reference with phase ϕ_-^{ref} . Another ion of identical species is confined in the Penning trap for an additional time $t_{acc} = t_{final} - t_0$ (accumulation time) and is ejected at t_{final} . During the accumulation time t_{acc} , the ion undergoes n_- full magnetron revolutions with radius r_- and angular frequency ω_- and accumulates a total magnetron phase of $\phi_-^{final} + 2\pi n_-$. The ion is thus ejected at time t_{final} and the detected ion spot has a phase angle ϕ_-^{final} (figure 3.6). The phase difference between the final and reference phases determines the magnetron frequency:

$$\nu_{-} = \frac{(\phi_{-}^{final} - \phi_{-}^{ref}) + 2\pi n_{-}}{2\pi t_{acc}}$$
(3.13)

The measurement scheme for the reduced cyclotron frequency differs slightly from the magnetron frequency measurement as shown in figure 3.7. In a real Penning trap, a residual axial motion can often not be avoided. Hence, the ions oscillate in the axial direction in addition to their radial motion and are ejected from different axial positions. This results in a time-of-flight distribution at the detector with a typical



Figure 3.7: Schematic depiction of the excitation-pulse scheme of the single-pattern PI-ICR measurement technique for the (a) reference phase and (b) final phase for independent measurements of the magnetron and reduced cyclotron frequencies. For the center spot projection, the ion is extracted without any excitation or phase accumulation.

spread of ~ 1 µs. During this time, an ion in a cyclotron motion of the order of a few MHz accumulates a larger phase, compared to one in a slow magnetron motion. Thus, just before ejection, an rf quadrupolar excitation at $\nu_{rf} = \nu_c$ is applied which converts the modified cyclotron motion to magnetron motion (step 2a in figure 3.7). In order to obtain the reference phase for the modified cyclotron spot, conversion is done immediately after dipolar excitation of the cyclotron motion. The ejection pulse is then applied and a phase angle ϕ_+^{ref} at time t_0 is obtained. For the determination of the accumulated phase after a storage time t_{acc} , the conversion is done after waiting for time t_{acc} , during which the ion has modified cyclotron motion only acquiring a $\phi_+^{final} + 2\pi n_+$ phase, such that:

$$\nu_{+} = \frac{(\phi_{+}^{final} - \phi_{+}^{ref}) + 2\pi n_{+}}{2\pi t_{acc}}.$$
(3.14)

It is important to note that the conversion pulse preserves the magnitude but changes the sign of the accumulated cyclotron phase [21, 92]. If the same conversion pulse is applied for both the reference and final phase measurements, the effect of conversion is the same for both of the phases and the phase difference remains unaffected. In order to project the center spot, no excitations are applied. If the same accumulation time is chosen for both the magnetron and the modified cyclotron frequency measurement, the cyclotron frequency is given as:

$$\nu_c \approx \nu_+ + \nu_- = \frac{\left(\left(\phi_+^{final} - \phi_+^{ref}\right) + \left(\phi_-^{final} - \phi_-^{ref}\right)\right) + 2\pi(n_+ + n_-)}{2\pi t_{acc}}.$$
 (3.15)

From the discussion above, it is evident that the measurement of four phases is required for the measurement of the cyclotron frequency of an ion species. In theory, a single ion is enough for the measurement of each phase and a minimum of five ions (including the center) would be sufficient for using the single-pattern PI-ICR technique. However, in reality, a larger number of ions is required in order to determine the phase correctly.

3.6.2Double-pattern scheme: direct measurement of ν_c

By measuring only the final phases of the modified cyclotron and the magnetron motions, a direct measurement of the free cyclotron frequency can be achieved with the double-pattern PI-ICR technique. Since only two phases need to be measured in the double-pattern scheme, the statistics required to perform a measurement is reduced by half in comparison to the single-pattern scheme. This makes the doublepattern PI-ICR technique ideal for experiments with low ion rates.

The measurement scheme is shown in figure 3.9. As in the case of the singlepattern scheme, a cooled ion is injected into the measurement trap. The ion is then subjected to a dipolar excitation at ν_+ , placing the ion in a cyclotron orbit as seen in step 2, figure 3.9. The major difference between both the magnetron and the cyclotron spots is at step 3. In order to project the magnetron phase, the ion is immediately exposed to a quadrupolar excitation to convert its motion entirely to the magnetron mode. The ion accumulates a magnetron phase during the time t_{acc} before extraction. However, in the case of the modified cyclotron spot, the ion is allowed to undergo cyclotron motion for t_{acc} before applying the conversion quadrupole pulse followed by an extraction pulse.

Figure 3.8: Projection of the magnetron and the modified cyclotron motion of ions in a Penning trap onto a position-sensitive detector. The trap-center image is obtained without any excitation. The magnetron and modified cyclotron spots correspond to radially excited ions after accumulating a phase during time t_{acc} [91].





Figure 3.9: Schematic depiction of the excitation-pulse scheme of the double-pattern PI-ICR measurement technique for direct measurement of the cyclotron frequency. Both magnetron and reduced cyclotron frequency measurements differ in the application of the conversion π -pulse at step 3. Note that the phase accumulation time t_{acc} in both cases is the same.

With n_{\pm} as the integer numbers of revolutions and ϕ_{\pm}^{final} as the relative phase angles built up in the radial motion during the time t_{acc} (figure 3.8), the frequencies are given as:

$$\nu_{\pm} = \frac{\mp (\phi_{\pm}^{final} - \phi_0) + 2\pi n_{\pm}}{2\pi t_{acc}}$$
(3.16)

and

$$\nu_c = \frac{(\phi_-^{final} - \phi_+^{final}) + 2\pi(n_+ + n_-)}{2\pi t_{acc}}$$
(3.17)

where ϕ_0 is the initial phase angle and is the same for both the magnetron and modified cyclotron spot.

The change of sign in the phase of the reduced cyclotron frequency in equation 3.16 can be attributed to the conversion quadrupole pulse which preserves the magnitude but flips the sign of the angle, as discussed in [21, 92]. The trap center in this case is also measured in the same way as in the single-pattern scheme, i.e. without any excitation.

For the measurements with nuclear isobars or high energy isomers, the concept of 'magnetron splitting' can be implemented. The magnetron spot for each isobar/isomer can be separated. This angle difference can be introduced between ion spots by adjusting the time t_s (figure 3.9). When no splitting is desired, t_s is small, and the conversion pulse with frequency ν_c is applied immediately after dipolar excitation at ν_+ . However, to introduce splitting between the spots, t_s is adjusted such that after excitation with ν_+ ions acquire slightly different modified cyclotron phases before the conversion pulse at ν_c is applied, thereby resulting in two different spots. After the conversion pulse, the ions accumulate the magnetron phase during the accumulation time (t_{acc}). This is defined as 'magnetron splitting' and is used in this work for the mass measurement of the isobaric mass doublet of ²⁰⁷Tl and ²⁰⁷Pb (section 7.3.6).

The position-sensitive delay-line detector at SHIPTRAP [93] is placed at a distance of 80 cm from the center of the measurement trap (section 6.5). It could not be placed any closer due to the restriction of maximum field strength of 100 mT at the detector. The detector magnifies the projection of the ion motion in the trap by a factor G, given by [20]:

$$G \approx \sqrt{\frac{B_z^{trap}}{B_z^{det}}} \tag{3.18}$$

where B_z^{trap} is the z-component of the magnetic field at the trap center (with z-axis as the symmetry axis of the magnetic field) and B_z^{det} is the z-component of the magnetic field at the detector. Equation 3.18 holds in the absence of an electric field and gives a value of $G \approx 20$ for the SHIPTRAP set-up.

3.7 Determination of atomic mass and mass excess

The mass of an ion can be determined from the measurement of its cyclotron frequency and magnetic field strength subjected upon it $(m = qB/2\pi\nu_c)$. In order to measure the atomic mass with high precision, it is required to have highly accurate cyclotron frequency and magnetic field strength measurements. This is achieved by taking the frequency measurements (using PI-ICR) of the ion of interest relative to a reference ion whose mass is known with at least the same precision as the ion of interest, defining the frequency ratio, R as:

$$R = \frac{\nu_c^{ref}}{\nu_c} \tag{3.19}$$

where ν_c^{ref} is the measured cyclotron frequency of the reference ion and ν_c is the cyclotron frequency of the ion of interest.

As explained in Section 7.2.4, the magnetic field has non-linear temporal variations such that the cyclotron frequency at a time t is defined as:

$$\nu_c(t) = \frac{1}{2\pi} \frac{qB(t)}{m} \tag{3.20}$$

The frequency ratio (equation 3.19) should ideally be obtained for the same time of measurement for both of the frequencies. However, in practice, this is not possible and the measurements of the reference ion alternate the measurements of the ion of interest. In the PI-ICR technique, the reference ion is measured at two different times t_i and t_{i+1} and the ion of interest is measured at the time t_j such that $t_i < t_j < t_{i+1}$. During the analysis the frequency for the reference ion is interpolated at the time of measurement of ion of interest.

The mass ratio is given by:

$$\frac{m_{exp}}{m_{ref}} = \frac{q}{q_{ref}} \frac{\nu_c^{ref}}{\nu_c} = \frac{q}{q_{ref}} R \tag{3.21}$$

where m_{ref} and m_{exp} are the masses of the ions corresponding to the reference and ion of interest. q is the charge on the ion of interest and q_{ref} is the charge of the reference ion. The experimentally obtained atomic mass for the desired element X $m(^{A}X_{Z})$ is:

$$m(^{A}X_{Z}) = R \frac{q}{q_{ref}}(m_{ref} - q_{ref}m_{e}) + qm_{e}$$
 (3.22)

where m_{ref} is the literature value of atomic mass of reference and m_e is the mass of electron. It is often more convenient to compare mass excess values which can be calculated as:

$$ME(^{A}X_{Z}) = (m(^{A}X_{Z}) - A)u$$
 [in keV/ c^{2}] (3.23)

where Z is the number of protons and A is the total number of protons and neutrons in the element X. Also, decay spectroscopy experiments allow for the measurement of the atomic mass of the ion of interest by the measurement of its decay energy. Table 3.1 lists all the reactions that contribute to the literature value related to the evaluation of the atomic mass of the elements discussed within the scope of this work. For the case of α -decay, the decay energy Q_{α} corresponds to the binding energy difference between the parent nucleus and its decay products (daughter nucleus and α particle). The nuclear equation describing α -decay is:

$${}^{A}\mathrm{X}_{Z} \rightarrow {}^{A-4}\mathrm{Y}_{Z-2} + {}^{4}\mathrm{He} + Q_{d}$$

The atomic mass of the nucleus X that decays via α -decay is given by:

$$m(^{A}X_{Z}) = m(^{A-4}Y_{Z-2}) + m(^{4}He) + Q_{\alpha}$$
 (3.24)

and the associated uncertainty δm_X to the mass determination is:

$$\delta m_X = \sqrt{(\delta m_Y)^2 + (\delta m_{He})^2 + (\delta Q_\alpha)^2}$$
 (3.25)

where $\delta m_i \forall i \in [Y,He]$ is the mass uncertainty of the respective elements obtained from AME 2020 [88] and δQ_{α} is the uncertainty in the measurement of decay energy (from the experiments). For the calculation of $m({}^{A}X_{Z})$, the mass values of the decay products are also taken from AME 2020. Hence, the absolute mass uncertainty of the atomic mass of ${}^{A}X_{Z}$ is thus limited by the imprecise knowledge of the mass of decay products.

Table 3.1: List of the mass equations for various reactions. The reactions contributing to the literature value from AME 2020 [88] for the masses measured within the scope of this work are listed in this table. In the second column, the formula to determine mass $m({}^{A}\mathbf{X}_{Z})$ from the respective decay energies is shown, followed by the uncertainty calculation in the third column. The mass excess for the comparison can thus be calculated using equation 3.23.

Reaction	Mass $m(^{A}X_{Z})$	Uncertainty $(\delta m_X)^2$
$^{A}\mathbf{X}_{Z}(\alpha)^{A-4}\mathbf{Y}_{Z-2}$	$m(^{A-4}Y_{Z-2}) + m(^{4}\text{He}) + Q_{\alpha}$	$(\delta m_Y)^2 + (\delta m_{He})^2 + (\delta Q_\alpha)^2$
$^{A}\mathbf{X}_{Z}(\beta^{-})^{A}\mathbf{Y}_{Z+1}$	$m(^{A}\mathbf{Y}_{Z+1}) + E_{\beta}$	$(\delta m_Y)^2 + (\delta E_\beta)^2$
$^{A}\mathbf{X}_{Z}(n,\gamma)^{A+1}\mathbf{Y}_{Z}$	$m(^{A+1}\mathbf{Y}_Z) - m(n) + E_\gamma$	$(\delta m_Y)^2 + (\delta m_n)^2 + (\delta E_\gamma)^2$
$^{A}\mathbf{X}_{Z}(d,p)^{A+1}\mathbf{Y}_{Z}$	$m(^{A+1}Y_Z) + m(p) - m(d) + m_e + E_{d,p}$	$(\delta m_Y)^2 + (\delta m_d)^2 + (\delta m_p)^2 +$
		$(\delta m_e)^2 + (\delta E_{d,p})^2$
$^{A}\mathbf{X}_{Z}(t,p)^{A+2}\mathbf{Y}_{Z}$	$m(^{A+2}Y_Z) + m(p) - m(t) + m_e + E_{t,p}$	$(\delta m_Y)^2 + (\delta m_t)^2 + (\delta m_p)^2 +$
		$(\delta m_e)^2 + (\delta E_{t,p})^2$

Chapter 4 The ARTEMIS experiment

The experiment is located at 'GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt' (GSI Helmholtz center for heavy ion research) and aims to measure the magnetic moment of electrons bound to heavy, highly charged ions. GSI is a heavy ion research facility established in 1969, in Darmstadt, Germany. The facility has accelerator structures which can provide nearly any ion of interest to various experiments. The stable lower charged ions are created at the 'Ion Source' and transferred to UNIversal Linear ACcelerator (UNILAC) [94], which is the heavy-ion injector for GSI. Highly energetic ions with an energy of 11.4 MeV/u from UNILAC are injected into heavy-ion synchrotron of GSI (SIS-18 or SchwerIonen Synchrotron) for acceleration [95]. These extremely energetic heavy, highly charged ions with peak energy of about 1 GeV/u are passed through thin stripper foils to further strip-off electrons and achieve the desired highly charged ionic states, as high as bare uranium. The ions extracted from SIS with approximate energy of 560 MeV/u are then sent to the Experimental Storage Ring (ESR), capable of storing the ion beams and decelerating to energies of 4 MeV/u.

In order to perform precision experiments, the ions need to be further cooled down. The ions extracted from ESR are transported to HITRAP (Heavy highly charged Ion TRAP). These ions are slowed down in two rf-decelerator structures to an energy of 6 keV/u for further cooling to a temperature as low as 4 K using resistive and electron gas sympathetic cooling at the HITRAP facility [96]. The ions having energy of 5 eV/u are then injected to the experiments connected to the HITRAP beamline, such as ARTEMIS and SPECTRAP. In order to successfully trap the ions coming from HITRAP beamline, ions need to have a sufficiently low energy (~ 100 eV/q). In ARTEMIS, this is achieved by implementation of a pulsed drift tube (details of the set-up in [97]). Figure 4.1 shows an overview of the GSI heavy ion research facility.

4. The ARTEMIS EXPERIMENT



Figure 4.1: Visualisation of GSI Helmholtz center for heavy ion research, Darmstadt. Courtesy: W. Geithner, GSI heavy ion research facility, 2017

The ARTEMIS experiment is connected to the HITRAP beamline (number 5: figure 4.1) and sits on top of ESR platform in the experimental hall (number 6).

4.1 The experimental set-up

ARTEMIS stands for the AsymmetRic Trap for the measurement of Electron Magnetic moment in IonS. The experiment has a uniform magnetic field of 7 T provided by a superconducting (NbTi) solenoid magnet coil from VARIAN. In order to avoid magnetic field distortions from the surroundings, the magnet cryostat is mounted on an aluminium ITEM frame surrounded by a wooden tower. The NbTi solenoid magnet is immersed in liquid helium and is cooled to 4 K. The 300 L helium vessel is surrounded by a 240 L liquid nitrogen dewar in order to provide an intermediate temperature range of 77 K. This prevents quick boil-off of helium and the tank lasts about 8-9 months with roughly 1% liquid boil-off every 3 days. The nitrogen vessel has to be filled once every 2 weeks due to a boil-off rate of 6-7% per day. The magnet was initially energised with a current of 210 A in 2009 and has been operational in a persistent mode since then. The magnetic field is homogeneous in a volume of about 1 cm³ with a specified homogeneity of better than 10^{-7} . The homogeneity at the center is better than 10^{-5} in a cylindrical volume of 0.5 cm diameter and a length of

10 cm. Such highly homogeneous field is achieved by the use of additional shimming coils [98].

The cryogenic part of the experiment consists of a gas injection system (replaced with the connection to HITRAP beamline [97]), the trap chamber, the electronic detection unit and the cryocooler (figure 4.2). The experimental set-up hangs into the 1730 mm long and 160 mm diameter 'warm-bore' of the magnet (bore at room temperature). The cryocooler is a two stage pulse-tube cooling unit SRP-82B from SUMITOMO HEAVY INDUSTRIES [99]. First stage with a cooling power of 40 W cools to a temperature of roughly 45 K, depending on the heat load on this stage. The second stage connects to the trap chamber and detection electronics through a long OFHC copper rod and has a cooling power of 1 W at 4 K. A long aluminium radiation shield is connected to the first stage in order to prevent radiative transfer of heat from the room temperature bore to the 4 K stage. Depending on the heat load, the temperature of the radiation shield can vary between 30 K and 45 K. In order to minimise this heat load, constantan wires insulated with teflon or PFA (perfluoroalkoxy alkane) are utilised in the experiment. The coaxial cables having teflon insulated brass inner wires and CuNi (copper nickle) outer shield are used for carrying the rf signals. The sapphire thermocouplers between the first and the second stage further ensure thermal insulation along with the electrical conduction for all cables going from room temperature to 4 K.

An inert gas (such as argon) is introduced into the ultra-high vacuum trap chamber using the gas injection system in order to create highly charged ions. These ions are created inside the trap using electron impact ionisation. Lately in February 2022, this has been replaced by a vertical connection to the HITRAP beamline for injection of heavy, highly charged ions produced through an external ion source [97]. The trap consists of a stack of cylindrical electrodes with a creation trap (CT) and a spectroscopy trap (ST). The pressure inside the trap chamber is better than 10^{-14} mbar (section 5.7). The motional frequencies of the created ions are detected non-destructively using resonators. These resonators are connected to the four-leg structure (figure 4.2) and together with the cryogenic filterboards form an electronic detection unit.

The whole experimental set-up along with the cold head and the radiation shield weighs over a 100 kg. When the experiment is lowered into the magnet bore, eddy currents are induced due to Lenz's law. These eddy currents exert a force on the magnet coil. This force should not exceed a value of 100 Newton. The experiment then rests on the bottom anti-cone part which is connected to the PEEK structures

4. The ARTEMIS EXPERIMENT



Figure 4.2: A CAD drawing of the set-up of the ARTEMIS experiment along with a magnified sectional view to show the technical details of the electronics, the gas-injection system and the trap.

(Poly Ether Ether Ketone), as shown in figure 4.2. These, together with a set of stranded copper wires connected to the four-leg structure, decouple the vibrations from the cryocooler and turbo pumps. After achieving a pressure of $\sim 10^{-5}$ mbar in the magnet bore using a turbo pump, the cryocooler is turned on in order to cool down the experiment. When the experiment is at a temperature of ~ 4 K, the cryocooling takes over and the turbo pump is not required anymore. Therefore, the valve at the top of the ARTEMIS tower connecting the experiment to an external rough pump can be closed and the turbo pump can be turned off to further reduce the vibrations.

Since there is no physical access to the experimental set-up once it is in the magnet bore, it has to be taken out every time any changes are foreseen. This requires bringing the experiment to atmospheric conditions of 300 K and 1000 mbar pressure before it is ready to be taken out of the bore. To take the experiment out, a two storey tall metal frame is set up on top of the tower with the help of a crane. This frame only stays up temporarily during the process of extracting or inserting the set-up from or into the magnet, respectively.

4.2 ARTEMIS Penning trap

The ARTEMIS trap stack is made of two connected Penning traps; the creation trap (CT) and the spectroscopy trap (ST) (figure 4.3). These hollow cylindrical electrodes in the Penning trap are made of OFHC (Oxygen-Free High thermal Conductivity) copper and have a 20 µm thick diffusion barrier, with less than one micrometer thick protective gold-plating layer. The electrodes with inner diameter of 17.513 mm are machined to micrometer precision. For electrical insulation, these electrodes are separated by 3 mm-thick sapphire spacer rings in the ST and 3 mm-thick macor spacer rings in the CT. The trap stack is held together by the six copper rods attached to the top and bottom of the trap and is bolted to the UMF ('Unterer Montageflansch') on the top, as shown in figure 4.3. The z-axis of the trap is aligned as parallel to the B-field axis as possible, in order to have minimal tilt between the two. A small tilt modifies the individual motional frequencies of the ions (ν_z , ν_+ and ν_-), but the true cyclotron frequency (ν_c) remains unaffected from these tilts due to the Brown-Gabrielse invariance theorem (equation 2.13). Electrodes E 8 to E 21 constitute the creation trap, which accounts for the in-flight capture and creation of ions. The spectroscopic measurements on the ion cloud are performed in the spectroscopy trap (electrodes E 1 to E 6). Electrode E 7 is the transport electrode which connects both these traps, so the ions can be transported by slow-adiabatic transfer between the traps. Table 4.1 depicts the geometrical parameters of both the creation trap and the spectroscopy trap. Both these traps are described in detail below.

Table 4.1: Geometric trap parameters for the ARTEMIS Penning traps: creation trap and spectroscopy trap. The coefficients are explained in section 2.1. The values for the trap coefficients are taken from [34].

	$z_0 \; [\mathrm{mm}]$	$\rho_0 \; [\mathrm{mm}]$	d [mm]	C_2	C_4	B[T]
Creation trap (E14)	7.459	8.704	6.7275	0.56305	0.001	7.000
Spectroscopy trap (E 3)	8.985	8.704	7.7075	0.52286	< 0.001	7.003

4.2.1 The creation trap

The creation trap at ARTEMIS is a mechanically compensated open-endcap trap (section 2.1.1). The trap is designed in such a way that it fulfills the requirements of *in-situ* creation of ions. It is also capable of in-flight capture and storage of ions from the external sources of ions, like EBITs or the HITRAP beamline. Alternating negative and positive voltages are applied to the 9 electrodes (E 10 to E 18). This



Figure 4.3: (a) Photograph of the ARTEMIS Penning trap when the trap chamber is removed showing the different electrodes and connections to the UMF. (b) A CAD drawing of the dual-Penning trap depicting different electrodes with their labels.

forms three consecutive harmonic traps where the ions can be trapped. The high voltage electrodes E 8, E 9 and E 21 confine the electron beam from the field emission point (FEP, E 19 electrode) and facilitate the in-flight capture of ions.

In the ion creation process using FEP, the amount of current depends on the voltages applied to the FEP and to the accelerator electrode (E 20). Since there are no compensation electrodes, thus the design of the trap electrodes for CT is done cautiously, such that the ring electrode follows $\rho_0 \approx 1.203 \ z_0$, thereby leading to $D_4 = 0$. The dimensions of electrodes in CT are $\rho_0 = 8.704 \text{ mm}, \ z_0 = 7.549 \text{ mm}$ which gives $d = 6.838 \text{ mm}, \ C_2 = 0.5631 \text{ and } C_4 = 0.001$. The electrodes E 10 to E 18 are connected to HV 250-8 from STAHL-ELECTRONICS which can supply $\pm 250 \text{ V}$ [100]. Therefore, this can form deep potential wells to store hot ion clouds with a large number of ions. An exception to this is the electrode E 14, the ring electrode, which is connected to the HV 500-8 from STAHL-ELECTRONICS to assist in cleaning of the ion ensemble using the SWIFT technique (stored waveform inverse Fourier transform, section 5.6). The created ion cloud is usually stored in E 14 and an axial resonator ARES CT is connected to E 13 (section 4.4). E 15 is a split electrode where the magnetron excitation can be applied (for magnetron cooling, see section 5.5) and



Figure 4.4: CAD drawing for the half-open spectroscopy trap indicating the optically transparent ITOcoated window with the large solid angle.

the axial excitation is applied at E16. Both CT and ST introduce different trap capacitance as load value to the resonators (ARES CT, ARES ST and CRES ST) connected to electrodes E13, E2 and E4, respectively (section 5.1).

4.2.2 The spectroscopy trap

The spectroscopy trap (ST) is a half-open electrically compensated trap (section 2.1.2). As the name suggests, the spectroscopic measurements on the trapped highly charged ions are performed in the ST. In order to perform high-precision spectroscopic and cyclotron frequency measurements on the ion cloud in ST, the trap is designed to be highly harmonic. As discussed in section 2.1, for a cylindrical Penning trap, the closed-endcap electrically compensated trap design gives the best harmonicity, but this would restrict the optical access to the trap. Hence, the next best option is to use the open-endcap design with long endcap electrodes. They simulate the potential of a closed-endcap design, meanwhile facilitating the required optical access. Even with the use of long endcap electrodes (with length roughly $4 z_0$ and $z_0 \simeq \rho_0$), the solid angle is 0.2 steradians. Thus, for the ARTEMIS spectroscopy trap, one of the endcaps is replaced with an electrically conducting and optically transparent Indium Tin Oxide window (ITO, section 4.2.3). ITO provides greater optical access with a solid angle of 2 steradian, thereby increasing the overall light collection efficiency. The other long endcap is replaced by a set of electrodes identical to the 'original' trap electrodes, i.e. the compensator and the ring electrodes but with opposite voltages applied to them. This forms an 'anti-trap' with anti-compensator and anti-ring electrodes (figure 4.3). The anti-trap simulates the potentials in such a way that the trap remains harmonic. The formation of consecutive short traps allows for the slow adiabatic transfer between the creation trap and the spectroscopy trap (which is not possible with a single long endcap electrode). The importance of the adiabatic transport process for cooling and ion ensemble preparation is discussed in section 5.4.

The rotating wall technique can be used to increase the ion density, hence bringing more ions in the focus of the laser resulting in increased fluorescence light from the spectroscopy. For this reason, the E3 electrode (ring electrode) is segmented into 4 parts which are electrically isolated by placing sapphire balls into the specially designed grooves. The axial resonator for ion detection is connected to E2 and the cyclotron resonator is connected to the split electrode E4. All the electrodes in ST have identical dimensions, except the spacing between E4 and E5 which is double the other gaps to have symmetric dimensions to the 'original' trap (figure 4.4). The electrodes in ST are biased using the HV 200-8 from STAHL ELECTRONICS which can provide $\pm 200 \text{ V}$, but due to the limitation from the filter board, the voltage limit is set at $\pm 65 \text{ V}$.

4.2.3 Optically transparent endcap window for the spectroscopy trap

The half-open Penning trap design provides highly-harmonic trap potentials in addition to excellent optical access to the ions. In order to have large optical access to the ions, a thin fine gold-plated mesh was used as an endcap electrode in the ST. The average light transmission from the mesh has a value of about 60% and it simulates potential of a closed surface. However, the peak laser beam can hit the strands of the mesh, leading to a loss in efficiency. Additionally, the non-planar surface of the mesh introduces anharmonicities in the trap potentials. Thus, the mesh was replaced by an optically transparent and electrically conducting Indium Tin Oxide coated glass window (ITO) forming optically open and electrically closed electrode. Indium Tin Oxide is a mixed oxide of indium and tin in varying proportions. Due to its electrical conductivity, it is also used as a Faraday cup, whereby destructively detecting the ion current created by the charged particles hitting the surface of the window. In ARTEMIS, the WTSQ11050-A window from THORLABS is used as an endcap electrode for the spectroscopy trap. It has an ITO coating on one side and anti-reflective coating for wavelengths of 350-500 nm on the other side of NBK-7 substrate window. The use of ITO windows in ARTEMIS increased the transmission for the 441 nm light (for Ar^{13+}) to 77%. Figure 4.5 shows the transmission for various materials used as for coating the endcap electrode [101].

For the spectroscopic measurements of heavy, highly charged ions such as hydrogenlike bismuth (Bi^{82+}), the hyperfine transitions are in the ultraviolet region, with a wavelength of roughly 243 nm (section 3.3). As can be seen from the figure 4.5, the transmission from the ITO is almost negligible below 250 nm. On the contrary,



Figure 4.5: Transmittance data of different transparent conductors: graphene, ITO, single-walled carbon nanotubes (SWNTs), ZnO/Ag/ZnO and TiO₂/Ag/TiO₂ for the endcap electrode [101]. Graphene and ITO are of special interest due to their high transmittance in the region of fine-and hyperfine transitions .

graphene offers exceptional transmittance over a wide range of spectrum. Therefore, it is a better option to be used as the endcap electrode to the ST in place of the ITO window and has more widespread applications [102].

As a substrate, materials such as CaF₂, MgF₂, Sapphire, α -BBO (alpha BaB₂O₄), BaF₂ and UV fused silica (UVFS) have excellent transmissions in UV regime and can be possible choice of substrate for the endcap window. On comparing the transmission data given by the 'THORLABS', the substrates UV fused silica, CaF₂ and BaF₂ can be used over a wider spread of wavelength with sufficiently good transmission. BaF₂ is avoided because of its hazardous nature and less resistance to water damages. Thus, a circular window of 25.4 mm diameter and 5 mm thickness, each of CaF₂ and UV fused silica, is taken and the transmission from both windows is tested using the 285 nm UV laser from LIBELLE experiment and observed to be 88.16% for UV fused silica and 99.14% for CaF₂. This data agrees to the value provided by the manufacturer to within 10% error. At the given wavelength, CaF₂ window has a better transmission than UV fused silica. This can be used as the substrate for the graphene-coated optically transparent endcap window.

4.3 In-situ ion creation

In the creation trap of ARTEMIS, electrodes E8, E9 (high voltage electrodes) and electrodes E 19 - E 21 assist in the production of ions. The possibility of ion creation in the trap generates opportunities to perform various measurements independent of the beamtime or EBITs' availability. In order to create highly charged ions (HCIs) in an EBIT, a thermionically emitted electron beam is utilised. However, this cannot be used at ARTEMIS because of the cryogenic environment. Hence, the electrons are produced by applying a high negative potential on a sharp tungsten needle 1 , the field emission point (FEP). This produces high electric field density at the tip due to which electrons are able to tunnel through the tip, resulting in the 'field emission'. For the ion creation, along with a high negative potential on the tungsten tip (FEP, electrode E 19), a relative positive potential is applied to the neighboring electrode called the 'accelerator electrode' (E 20) and a very high negative potential applied to the 'reflector electrode' (E21). The high voltages at ARTEMIS are applied using the high voltage power supply HV-FEP from STAHL ELECTRONICS. This high voltage power supply can provide a potential of $\pm 3 \,\mathrm{kV}$ and measure current on the FEP, accelerator and reflector electrodes.

The highest charge state that can be achieved during creation depends on the electron energy which in turn is defined by the voltage applied to the FEP and the duration for which the voltage is applied. The potential on the accelerator electrode together with the shape of the tip determine the electron current density. A negative potential on the reflector electrodes (E 8, E 9 on one end and E 21 on the other) allows 'reuse' of the electrons by trapping them axially and the magnetic field provides radial confinement and facilitates the creation of highly charged ions.

In order to create argon ions, electron beam is produced using the FEP electron gun and is trapped between the high voltage electrodes and reflector electrodes. Neutral argon gas is injected into the trap by heating a cryogenic gas valve (Flotte-Lotte). The gas pressure and flow rate can be controlled by using the dosing valves in the injection system. Alternate positive and negative potentials are applied on the 9 trap electrodes of the CT, forming 3 consecutive traps. The electron beam hits the argon atoms and strips the electrons, creating positive ions. Since the electrons are trapped radially and axially, the 'charge breeding' continues further stripping electrons from the positive argon ions to form higher charge states. These positive ions are then

¹The tungsten needle is actually not a single rod, rather a combination of several tiny tungsten rods combining to form a 'tip' of the order of tens of nanometers. The tip has been characterised and tested in [34].


Figure 4.6: The cryogenic gas valve and ARTEMIS gas injection system. (a) A CAD drawing of the cryogenic gas injection valve. A heating resistor is placed on the copper structure (heater 1) and the stainless steel tube is wrapped with the heater wire (heater 2) to facilitate gas injection into the trap chamber. (b) The gas injection system at ARTEMIS. The two dosing valves control the argon gas flow and pressure in the injection line through the vacuum pump attached to the system. The duration for which the gas is injected is regulated by the pressurised air-valve followed by the manual valve. The outer injection line is attached to the steel tube connecting to the cryogenic valve through SWAGELOCK connector.

trapped in the 3 potential wells of CT. By changing the voltages applied to the electrodes, the ions from all 3 traps are concentrated into a single trap with E 14 as ring electrode. The created ions are then detected and identified using the resonators (explained in section 4.4).

To facilitate the *in-situ* ion creation along with the separation of the ultra-high vacuum conditions in the trap from the near atmospheric pressure of argon gas in the injection system, the cryogenic valve plays a very significant role. The cryogenic valve is a copper block with baffles cooled to cryogenic temperature (<20 K). At such a low temperature, gas atoms freeze to the surface of the baffles, thereby 'closing' the valve. In order to inject the argon gas into the trap, the heating resistors (heaters 1 and 2 in figure 4.6a) attached to the valve are heated for a few seconds. This elevates the temperature of the valve to ~30 K, thus 'opening' the valve for argon gas injection. At this point, the pressurised air-valve is opened and argon gas is injected into the trap. As soon as heating for the cryogenic valve is turned off, the temperature drops rapidly thereby sealing the valve again. More details on the ion creation system used in ARTEMIS and cryogenic gas valve has been provided in [103], [34] and [43].

4.4 Development and upgrade of non-destructive electronic detection system

As described in sections 2.5.3, detection and cooling are two different aspects of the non-destructive electronic detectors implemented in the form of 'resonators' at ARTEMIS. When the ions' motional frequency comes into resonance with the central frequency of the resonator, a signal is observed (equation 2.44). Since small amount of current (few fA to pA) is induced by the oscillating charged particles, cryogenic and room temperature amplifiers [43] are implemented in the detection system. Additionally, the varactor diodes boards, whose capacitance can be changed by an applied voltage, are used to have a tunable frequency range for the resonance frequency of the detection circuit (e.g. in the reduced cyclotron frequency detection at ARTEMIS). Therefore, the non-destructive ion motion detection system at ARTEMIS has three major components:

- 1. Resonator coils
- 2. Amplifier boards
- 3. Varactor diode boards.

The creation trap has one axial detection system (ARES CT) connected to E 13, while the spectroscopy trap has two resonators connected to it: the axial resonator (ARES ST) connected to E 2 while the detection system connected to E 4 (CRES ST) to measure the reduced cyclotron frequency (see figure 4.3 for details of the trap electrodes). In the following text, the resonator for reduced cyclotron frequency detection (ν_+) is termed as 'cyclotron resonator'. This section describes the developments and upgrades made to the detection system (resonators and amplifier board). Although no notable modifications have been performed for the varactor diode, they are included here, since they play an important role in the detection circuit and have been utilised for the characterisation of the load trap capacitance (section 5.1).

4.4.1 Resonator coils

For highly sensitive detection, the detection circuit should have a high Q-factor, minimum parasitic capacitance and high resonance resistance R_p (section 2.5.3). The resonators in ARTEMIS are self-wound coils enclosed in a conducting housing. These coils are fabricated by winding a wire on an insulator core, usually teffon. While designing the resonators various factors such as space availability, desired resonance



Figure 4.7: The CAD drawing of the cross-section of a helical resonator used in ARTEMIS. l_1 is the length of the windings, l_2 and D_2 is, respectively, the length and the internal diameter of the resonator housing.

frequency, cryogenic environment and magnetic field should be kept in mind. Because of the space limitations and frequency requirements at ARTEMIS, multi-layer helical resonators are fabricated for the axial frequency measurement. Various efforts have been made to optimise the resonator's design by using the semi-empirical formulas from [104, 105].

For a single-layer helical resonator with air-core, with a coil l_1 in length and having a diameter of D_1 (figure 4.7), the inductance is given by:

$$L \approx \frac{D_1^2 \times N^2}{18D_1 + 40l_1} , \qquad (4.1)$$

here D_1 and l_1 are in inches (figure 4.7) and the inductance is given in μ H. Using the equation above, an estimate on the number of turns can be obtained. For a resonator with housing diameter D_2 and length l_2 , the inductance is modified to be:

$$L_H \approx L \left[1 - \left(\frac{D_1}{D_2}\right)^3 \right] \left[1 - \left(\frac{l_1}{2l_2}\right)^2 \right]$$
(4.2)

Since the axial resonators need to be around the frequency of 1 MHz, the coil has about 250-300 turns. Based on the available space at ARTEMIS, a single long layer which accommodates all the turns could not be used. Hence, multiple layer resonator coils are built. In such a case, the inductance of the coil is calculated as:

$$L \approx \frac{0.2 \left(D_1^2 \times N^2\right)}{3D_1 + 9l_1 + 10D_w}.$$
(4.3)

Meanwhile, the capacitance for a single-layer air-core is estimated from turn-to-turn (C_{tt}) and turn-to-housing (C_{th}) contributions. For a wire of thickness r, with p

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Figure 4.8: Inductor tapping of the resonator coil in ARTEMIS. The tapping is done at 30% of the coil length from the ground (base) plane, with the shorter length being the secondary coil (shown in red) and the longer is the primary coil (shown in blue).



distance between two adjacent turns:

$$C_{tt} \approx \frac{\pi^2 \epsilon_0 D_1}{\ln\left(p/2r + \sqrt{(p/2r)^2 - 1}\right)} \tag{4.4}$$

and for a distance h of the outer coil layer from the housing:

$$C_{th} \approx \frac{2\pi^2 \epsilon_0 D_1}{\ln\left(h/r + \sqrt{(h/r)^2 - 1}\right)}.$$
(4.5)

where ϵ_0 is the absolute permittivity of free space. Here, all the dimensions are in meters and the capacitance is typically of the order of pF. The total capacitance C for an N-turn resonator is thus given by:

$$C = \frac{C_{tt}}{N} + \frac{C_{th}}{2} \tag{4.6}$$

Apart from these two contributions, other factors such as capacitance between adjacent layers also contribute in the case of multi-layer resonator. Since even the formulas for a single layer coil do not provide a good estimate on the value of capacitance, hence capacitance for multi-layer case was not investigated further. From these semi-empirical formulas discussed above, an estimate is made about the number of turns and inductance of the coil. However, it has also been observed that the attempts to accurately formulate the design parameters from these formulas were not entirely successful and the production of the resonators is based primarily on trial and error.

In order to decouple the external noise, inductor tapping is used in the ARTEMIS resonator coils by soldering a wire at a particular length of the coil. This creates a transformer [106] with a primary and a secondary coil as shown in figure 4.8 and is known as 'inductor tapping' or simply 'tapping'. This is a commonly used technique in the community to decouple the noise [58, 107]. The primary part is the longer

segment of the coil with $N_{primary}$ turns and is connected to the trap electrode through the 'hot end'. The secondary coil is the shorter part of the coil with $N_{secondary}$ turns. It is connected to the ground through the base plate of the resonator. The voltages across these two components of the transformer are related as:

$$\frac{V_{primary}}{V_{secondary}} = \frac{N_{primary}}{N_{secondary}};$$
(4.7)

where $V_{primary}$ and $V_{secondary}$ are the respective voltages across primary and secondary coils. In ARTEMIS, the tapping is done with 30% of total turns creating a secondary coil. However, it has been observed that due to the inductor tapping in the case of axial resonators, a dual peak structure is observed in the frequency spectrum of the resonator at the spectrum analyser. This has been explained later in this section. The point where the tapping is done is called the tap end and is used to input the signal from the resonator into the amplifier board. At the input of the amplifier board, the signal coming from the resonator is capacitatively coupled through a built-in high quality capacitor. Thus, both inductive and capacitative coupling techniques are used at different stages to reduce the external noise.



Figure 4.9: Steps for winding a helical resonator coil at ARTEMIS. The resonator has two layers of insulated copper wire separated by a layer of teflon tape.

The first step in making these coils is to ensure secure grounding of the wire used for winding the coil (figure 4.9). For this reason, the wire is soldered to the base plate of the resonator. The base and housing of the resonator are made-up of oxygen-free high-conductivity copper (OFHC). The axial coils are wound using 0.08 mm diameter copper wire with teflon insulation from OMEGA (IEC-TFCP-003). For the cyclotron resonators, an uninsulated copper wire with a diameter of 1 mm from GOODFELLOW is used. When one-thirds of the expected total number of turns are wrapped around the teflon core, the 'tapping' is done by soldering another wire to the coil. In order to make the tapped connection secure and to make the entire coil using a single piece

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Figure 4.10: CAD drawings for (a) helical and (b) toroidal resonators. The helical resonator has grooved-core structure in order to facilitate thermalisation for the superconducting wire. The axial resonators used in ARTEMIS with the normal conducting copper wire is without a grooved core.

of wire, a small capillary tube is added to the wire roll before soldering it to the base plate. After completion of each layer, teflon tape is wrapped around it to fix the windings and to provide a smoother surface for the next layer. The properties and geometric description of the three resonators currently used in ARTEMIS are shown in table 4.2. Since the resonators do not retain their Q-factor after several thermal cyclings, a new set of resonators should be fabricated each time the trap is taken out for modifications. Figure 4.9 depicts the winding process for a helical axial resonator with the copper wire of 0.08 mm diameter.

The teflon core of the resonator can be modified to have grooves, as in the case of cyclotron resonator at ARTEMIS, so the coil sits perfectly in the grooves and is rigidly held. The groove structure can also be applied for the superconducting coils to facilitate thermalisation of the core by increasing the contact area with teflon. Figure 4.10 shows the CAD drawings for the two different geometries of resonator. As discussed above, the helical resonators have been implemented in the ARTEMIS set-up due to space restrictions.

During the winding process of the resonator coil, the following points should be considered:

- 1. The grounding of the resonator wire should be ensured by heating the base plate and securing the wire using soldering material and a capillary tube.
- 2. The resonator housing and the base plate should be polished and cleaned to



Figure 4.11: Comparison of Q-factor for a coil made from copper wire with ~ 250 turns at room temperature and at 4 K. The quality factor increases multiple folds upon cooling to cryogenic temperature.

get rid of all the grease. The resonator housing is polished thoroughly using a metal polish and then cleaned with isopropanol. This is specially important in helical resonators, since the field line penetration onto the resonator housing is not zero. Hence, these stray effects can reduce the quality of the resonator.

- 3. It should be ensured that the whole coil should be made with a single continuous wire to avoid any unknown capacitance creeping in at the joint. This is also to avoid any cracking of solder when cooled to 4 K, thus rendering the coil useless.
- 4. The spacing between the turns during the coil winding should be as uniform as possible. A variation of distance between two consecutive turns would lead to a change in the capacitance value associated with it (equation 4.4).

After winding the coil, the resonators are examined in two different configurations a) without connecting an external load capacitance to know the frequency of resonance of the coil and b) by using different values of high quality capacitors as load to determine the self capacitance and inductance of the coil (using equation 2.41). The tests are performed using E5080A ENA network analyser from KEYSIGHT. Before installation of the detection system in the ARTEMIS experimental set-up, the resonators are tested at the cryogenic test-stand (appendix A).

The quality factor of the inductor coil made with a copper wire increases multiple folds upon cooling it down to 4 K. The effect can be seen in figure 4.11, where a helical coil with ~ 250 turns is observed with the network analyser at the test-stand. The measurements are performed at room temperature and at 4 K and more than ten fold increase in the quality is obtained. The change in noise level is due to the reduced thermal noise at a lower temperature.

All the three resonators installed in ARTEMIS are designed with a housing made of OFHC with outer diameter of 44 mm and thickness of 1 mm. The housings are

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Table 4.2: Properties and geometric description of the axial and cyclotron resonators used at ARTEMIS during the measurement campaign in 2021. All the three resonators have helical teflon cores and the coils are made using copper wire. The resonance frequency at 4 K corresponds to the value of frequency obtained for the resonators in the ARTEMIS trap set-up by utilising a particular setting of the biasing voltages on the cryogenic amplifier boards (given in table 4.3).

	ARES CT	ARES ST	CRES ST	
Trap Electrode	E 13	E2	${\rm E}4$	
Design frequency at room temp. ²	$1.05\mathrm{MHz}$	$890\mathrm{kHz}$	$102.3\mathrm{MHz}$	
Q-factor at room temp. ²	50	50	116	
Resonance frequency at 4 K $(\nu_0)^3$	$705.7\mathrm{kHz}$	$746.7\mathrm{kHz}$	$33.1\mathrm{MHz^4}$	
Q-factor in ARTEMIS set-up ³	376	345	33^{4}	
Wire diameter	$0.08\mathrm{mm}$	$0.08\mathrm{mm}$	$0.75\mathrm{mm}$	
Insulation thickness	$0.04\mathrm{mm}$	$0.04\mathrm{mm}$	No insulation	
Number of windings	${\sim}245~{\rm turns}$	${\sim}280~{\rm turns}$	$6.3 \mathrm{turns}$	
Self capacitance of resonator	$32.5\mathrm{pF}$	$26.6\mathrm{pF}$	$4\mathrm{pF}$	
Inductance of resonator	$0.7\mathrm{mH}$	$1.1\mathrm{mH}$	$75\mu\mathrm{H}$	

each 52 mm long. Also, the teflon core is designed such that it remains in contact with the resonator body on both ends to ensure good thermal contact.

As depicted in table 4.2, both the axial resonators are designed to have a resonance frequency of ~ 700-750 kHz. Since it is possible to scan the voltage to change the trap depth, ions with different charge-to-mass ratios come into resonance at this frequency (explained in detail in section 5.3). For the creation trap with an axial resonator having a central frequency of ~705 kHz, argon ions with charge states 2+ to 16+ can be observed in the trap. However, for the spectroscopy trap with an axial resonator at ~705 kHz, argon charge states ranging from 9+ to 16+ can be detected. The difference in the ions that come in resonance in both CT and ST is because of the voltage limitation of 65 V for the spectroscopy trap. The cyclotron resonator is designed to be operated at a frequency of ~35 MHz which is the reduced cyclotron frequency of Ar¹³⁺ ions in magnetic field of 7 T. The resonance frequency of the cyclotron resonator can be tuned by using a varactor diode board, as described in section 4.4.3. It should be noted that extremely high quality factors (Q = 10000 - 20000) are desirable for

 $^{^2\}mathrm{Design}$ values at room temperature tested outside the ARTEMIS experimental set-up (at the test-stand).

³Resonator installed in the ARTEMIS set-up. The load capacitance of the trap is the reason for the change in central frequency of the resonator

⁴The value corresponds to the frequency at 4 K in the trap with varactor diode connected to it such that the frequency can be tuned to match the reduced cyclotron frequency for Ar^{13+} ions.

detection but not for the cooling of large number of ions, as in ARTEMIS. The values of the Q-factor of the coil increase multiple folds upon cooling in the ARTEMIS setup⁵. The value of the design frequency of the resonator at room temperature is much higher than the value at 4K. This is because the measurements corresponding to the room temperature are taken at the test-stand (appendix A) without connecting to any load capacitance, however the 4K values in table 4.2 are with the resonators mounted to the trap thereby having the trap capacitance as load. Characterisation of the effect of the trap capacitance on the resonance frequency, signal-to-noise ratio and quality factor of the detection system is discussed in section 5.1.

Observation of two peaks in the frequency spectrum

It has been observed while winding the coil for the axial resonator that the frequency spectrum obtained on the spectrum analyser has two peaks in the region of interest. Observation of multiple peaks is not uncommon with the peaks appearing as higher harmonics of the coil's central frequency. However, the multiple peaks detected from the coils fabricated within the scope of this work are not higher harmonic peaks. Figure 4.12 shows the two-peak structure as observed in the resonance spectrum of one such coil, with the first peak at a frequency of 1.04 MHz and the second peak at f = 3.41 MHz.



Figure 4.12: Observation of two peaks in the frequency spectrum for a helical copper coil designed for the axial frequency measurements.

In order to understand the dual-peak structure, load capacitors with different values were connected in parallel to the hot end as well as in parallel to the tap end. Because of the presence of load capacitance (C_{load}) , the central frequency is observed to shift as expected from $f = \sqrt{1/(2\pi L(C + C_{load}))}$. However, the shift in frequency

 $^{{}^{5}}$ Except of the cyclotron resonator. This is considered to be a consequence of the presence of varactor diode and is further detailed in section 4.4.3



Figure 4.13: Variation of $1/f^2$ versus different load capacitance values for the observed two-peak structure. The load capacitance is applied in parallel to the hot end and the tap end.

is not the same for both peaks, indicating that the multiple peaks are not higher harmonics of the first observed peak. The graph for $1/\nu^2$ vs the load capacitance is plotted for both of the peaks with the load capacitance at hot end and at the tap end.

From the graph shown in figure 4.13, the self capacitance (C_p) and self inductance (L_p) value of the coil are extracted from the slope of the linear fit and y-intercept values according to the equations:

$$C_p = \frac{\text{intercept on y-axis}}{\text{slope of the linear fit}}, \quad L_p = \frac{\text{slope of the linear fit}}{4\pi^2}.$$
 (4.8)

In addition to this, the inductance of the coil between the hot end and ground, and between the tap end and ground are measured using an LCR meter. These values are compared to the corresponding inductance values obtained from the graph and the following conclusions were made:

- For the first peak, when the load capacitor is connected in parallel at the hot end, the value of the measured and the calculated inductance agree with each other ($L_{graph} = 655.9 \,\mu\text{H}$ and $L_{meas} = 708.1 \,\mu\text{H}$). However, this is not true when the load capacitor is connected at the tap end. This further implies, that the first peak corresponds to the oscillator formed by the windings between the hot end and ground .
- For the second peak, when the load capacitor is connected in parallel at the tap end, the value of the measured and the calculated inductance agree rather well with each other ($L_{graph} = 60.3 \,\mu\text{H}$ and $L_{meas} = 71.6 \,\mu\text{H}$). There is a strong disagreement in the values when the load capacitor is connected in parallel at

the hot end. Thus the windings between the tap end and the ground act as an individual oscillator with the resonance frequency at f = 3.41 MHz.

Thus, the first peak is considered as the central frequency of the resonator. Since the measurements on the ion cloud are made in the 'zero-span' mode of the spectrum analyser (section 5.3), the presence of second peak does not interfere with the measurements.

4.4.2 Cryogenic amplifier boards

The oscillating charged particles in a Penning trap create oscillating image currents on the electrodes. These image charges are usually small, on the order of a few fA to pA and hence, the signal needs to be amplified in order to detect them. Thus, the low-noise cryogenic amplifiers are an important part of the non-destructive detection system of ARTEMIS. The signal from the resonator coil is capacitatively coupled to the input of the cryogenic amplifier. The amplifiers used in ARTEMIS should have a high input impedance and low noise in order to sufficiently amplify the image current.

The cryogenic amplifiers used in ARTEMIS have two stages: the amplification stage and the impedance matching stage. The first stage, which is responsible for amplification of the signal uses a common source FET circuit and gives high voltage gain at high impedance. The second stage is a source follower FET circuit and matches the high output impedance of the amplification stage to the 50 Ω impedance of the rf system. Since GaAs is functional at cryogenic temperatures [108, 109, 110], the amplifiers are based on dual-gate GaAs metal semiconductor field effect transistors (MESFET).

MESFETs are active semiconductor devices with metal-semiconductor junction (Schottky junction) at the gate. The MESFET depicted in figure 4.14 is fabricated



Figure 4.14: Schematic drawing of the n-channel single-gate MESFET device [43].

on GaAs substrate with a lightly doped n-type semiconductor channel. To enable an ohmic contact with the external circuit, a highly doped semiconductor layers are grown which act as source and drain. The drain to source current can be controlled by changing the gate voltage which in turn changes the thickness of the depletion layer. MESFETs are used as low-noise power amplifiers in the millimeter and microwave range, but can also be used in the desired frequency ranges of kHz and MHz for the present application.

The amplifier boards used in the current set-up are based on the design devised by BASE collaboration at CERN [43, 58]. The amplifier boards mounted on all the three resonators currently used in ARTEMIS are fabricated within the scope of this work after implementing the changes listed in this section. In order to reach a high Q-factor value for the detection system, the losses in amplifier board such as the parasitic reactive loads, resistive losses and dielectric losses $(\tan \delta)$ should be minimised. The dielectric losses occur due to the polarisation current in dielectric materials. By optimising the circuit board design and substrate material, parasitic reactive components and dielectric losses can be reduced. Designing high input impedance amplifiers is important to lower the resistive losses in the system. The schematic design of the amplifier board used presently in the set-up is shown in figure 4.15.

The amplifier board is equipped using surface mount device (SMD) type components with 0603 package, except for the 1 μ F capacitor which has 1206 packages. The first stage is built around SONY 3SK164 'I' dual-gate transistors and the sourcefollower second stage is centered around SIEMENS CF739 'MSs' transistor. The amplifiers are to be operated in a cryogenic environment of 4 K and in a magnetic field of 7 T. For the selection of components to be mounted on the amplifier boards and for fabrication process, the following points are recommended⁶:

- 1. The circuit board material should have low dielectric loss tangent $(\tan \delta)$ and low thermal expansion coefficient. A good option for the circuit board material are teflon, sapphire and FR4 with $\tan \delta$ of 10^{-4} , 10^{-5} and 10^{-2} , respectively.
- 2. The solder wire should be compatible to the cryogenic temperatures in order to avoid cracking of the soldered joint upon cooling down to 4 K. For this reason, a special cryo-solder wire having antimony, tin and lead should be used.

 $^{^6\}mathrm{Based}$ on the discussions with S. Stahl from STAHL ELECTRONICS, and the trial and error process.



Figure 4.15: Schematic design of the amplifier board used at ARTEMIS. The coupling capacitor C1 couples the output from the resonator to the input of the amplifier board and can be varied in order to optimise the signal-to-noise ratio and the quality factor of the detection system.

- 3. Thick-film resistors are observed to have higher change in resistance values with a variation in temperature. Thin-film resistors from SUSUMU are used for making the amplifier boards.
- 4. It is recommended to use 'vintage' transistors, since they have more stable output.
- 5. The ceramic capacitors with 'X7R' series have higher series resistance, and their value change by $\sim 97\%$ when cooled. The JOHANSON (S-series) with high *Q*-factor, and 'CGA' capacitors are good alternatives. In order to avoid exposing the capacitors to higher temperatures, they should be the last component to be soldered on the board.
- 6. In order to ensure proper grounding of the amplifier board, the circuit board is soldered onto a copper plate. This conducting wires should be passed through 'vias' which are spread throughout the board. The 'vias' are the tiny tubes joining the top and bottom of the circuit board to ensure a proper contact to the ground.

7. The temperature on the soldering iron can be as high as 350-380 °C but the contact with the heated tip should not be longer than ten seconds.

During the measurement campaign of 2019, it was observed that the fabricated amplifier boards are not reliable after a few thermal cyclings. A number of factors were implemented during making the presently used amplifier boards, starting with the board material. The teflon boards were replaced with slightly larger FR4 boards in 2020 which are still being reliably used in the set-up. Despite higher dielectric losses, FR4 boards are found to function better due to their low thermal expansion coefficient $(10^{-5} \,\mathrm{K}^{-1}$ as compared to $10^{-4} \,\mathrm{K}^{-1}$ for teflon). As discussed above, the cryogenic antimony based solder is used and 100 M Ω resistors are replaced with thin-film resistors.

After fabrication of the filter board, it is investigated with the help of a microscope to ensure that there are no cracks in the soldered joints. To further improve the quality of the board and ensure stability of joints upon cooling, reflow soldering is done using RMA-23-UV flux from AMTECH ELECTRONICS. After heating with a heat gun, the excess flux material is removed using isopropanol to evade any unwanted conducting paths. This has significantly improved the durability of the cryogenic amplifier boards.



Figure 4.16: (a) Photograph of the amplifier board in the modified configuration. The amplifier board is soldered onto a copper plate which is mounted on the resonator housing with the help of a mounting adapter. (b) Photograph of the resonator with the modified configuration presently installed at ARTEMIS.

Another major variation adapted in 2021 measurement campaign is the position where the amplifier boards are mounted. As reported in the thesis of S. Ebrahimi [43], the amplifier boards were mounted on top of the resonators. In this orientation, the drain to source current was perpendicular to the magnetic field direction. It was observed that some of the transistors stopped functioning after they were placed in the cryogenic strong B-field environment of the trap. This could be attributed to quantum Hall effect. Hence, in 2021 the amplifier boards were mounted on the resonator housing (see figure 4.16) which in turn causes the drain-to-source current to be aligned along the magnetic field axis. Another advantage of the present orientation is the availability of larger area to mount the amplifier. This allows the possibility of having larger circuit boards which facilitate better grounding and heat sink. Aforementioned changes have helped to increase the reliability and longevity of the amplifier boards.

The next task after fabrication of the amplifier board is to find optimum parameters in order to bias the transistors for maximum possible quality factor and signal-tonoise ratio. As can be seen in figure 4.15, the amplifier boards used at ARTEMIS can be biased by using 5 channels, G 1, G 2, D 1 belonging to the first amplification stage, and G and D 2 to the second impedance matching stage. Because of the limitation on the number of biasing channels available in the set-up and the possibility of providing a single value of biasing voltage for both of the drain channels, each of the amplifier boards is biased using three input lines. The drains D 1 and D 2 are combined on a single channel and are biased with voltage V_D . Gate 1 for the first stage is biased using a voltage V_{G1} and gate 2 is connected to ground. The third biasing channel provides a voltage V_G to the gate of second transistor. Figure 4.17 shows the variation of quality factor and signal-to-noise ratio (SNR) for one such amplifier board in an attempt to find optimum voltage values. The term 'signal' here refers to the thermal noise from the electronic detection circuit above the background 'noise' level. The



Figure 4.17: Determination of the optimum biasing voltage for a two-stage amplifier. As the voltages on the drains and gates in the amplifier are varied, the Q-factor and the SNR change. The operational values for biasing voltage of the amplifier are found to be: $V_D = 3.2 \text{ V}$, $V_G = -0.75 \text{ V}$ and $V_{G1} = -1.75 \text{ V}$.

	ARES CT	ARES ST	CRES ST
Trap Electrode	E 13	E 2	E 4
V_D [V]	3.0	3.289	3.077
V_G [V]	-0.577	-0.569	-0.578
V_{G1} [V]	-1.953	-1.845	-1.92
$I_D [\mathrm{mA}]$	4.03	4.23	5.121

Table 4.3: Biasing voltages for three amplifiers corresponding to the three detection systems in ARTEMIS.

measurements are performed by varying one of the voltages for a constant value of other two biasing voltages and observing the corresponding Q-factor and SNR (e.g. to find optimum V_D , the values of V_G and V_{G1} are kept constant). It can be seen that the trend of variation of Q-factor is opposite to that of signal-to-noise ratio. Hence, for the data depicted here, the optimum biasing set is: $V_D = 3.2 \text{ V}, V_G = -0.75 \text{ V}$ and $V_{G1} = -1.75 \text{ V}$.

Table 4.3 gives the biasing voltages for each of the amplifier boards used in ARTEMIS measured at room temperature. These need to be fine-tuned slightly after the experiment is cooled down to the cryogenic temperature.

4.4.3 Varactor diode board

In an effort to measure the reduced cyclotron frequency using the cyclotron resonator, the resonance frequency of the detection system should be equal to motional frequency of the ion (ν_+). In a magnetic field of 7 T at ARTEMIS, Ar¹³⁺ ions have $\nu_+ \approx 35$ MHz. Aiming for high-precision g-factor measurement requires a precise knowledge of this frequency. Hence, a tunable capacitance is introduced in the cyclotron detection system in the form of a varactor diode board. The core of this varactor diode is a P-N diode whose capacitance and series resistance decreases with increase in the reverse bias voltage (U_{var}) [111]. Upon implementing this tunable capacitor, the total capacitance and quality factor of the detection system can be written as:

$$C(U_{var}) = C_{sys} + C_{var}(U_{var}) , \qquad \frac{1}{Q(U_{var})} = \frac{1}{Q_{sys}} + \frac{1}{Q_{var}(U_{var})}$$
(4.9)

where C_{sys} and Q_{sys} is, respectively, the capacitance and quality factor of the entire detection system except the varactor diode board with capacitance $C_{var}(U_{var})$ and quality factor $Q_{var}(U_{var})$.

Tests were performed by M. Wiesel [103] in order to check the compatibility of such a varactor diode in the high B-field cryogenic environment of ARTEMIS. These tests were done at the HILITE experimental set-up with varying magnetic field (up to 6 T) and temperature. It was concluded that in the presence of a B = 7 T the varactor diode can function only at a temperature greater than 20 K. Thus, a heatable varactor diode board was fabricated at STAHL-ELECTRONICS by using a MACOM MA46H202 varactor diode. The board has a nominal tuning range of 6 to 1.3 pF for the biasing voltage from 1 to 10 V. Figure 4.18 displays the schematic overview of the varactor diode board used in ARTEMIS. The board is grounded through the screws that connect it to the experimental set-up.

By changing the biasing voltage from 0-10 V on the varactor diode, the resonance frequency of the cyclotron resonator can be varied over a span of ~ 3.5 MHz. This is sufficient to provide the necessary flexibility in the frequency. As the biasing voltage increases, the capacitance decreases and hence the quality factor is expected to increase as

$$Q_{var}(U_{var}) = 1/2\pi f R_{var} C_{var}(U_{var}).$$

This can be seen in figure 4.19. The graph depicts the variation of resonance frequency, quality factor and signal-to-noise ratio with the change in biasing voltage. A table with the exact values corresponding to these measurements is included in appendix B. It should be noted that the capacitance shown in the figure does not correspond directly to the capacitance introduced by the varactor diode, rather to the capacitance of the system. Since all other components remain the same during the measurement, it can be regarded as a good metric for the capacitance of the varactor. The measurements shown here are done with the varactor heater set of 2 V and heating power of 200μ W, which is expected to raise the temperature of the varactor board optimum for its operation.

Since the trap voltage of CT and ST can be varied to bring different charge species into resonance at the axial frequency, the varactor diode is implemented only



Figure 4.18: Electronic layout of the heatable varactor diode board at ARTEMIS. The board is fabricated at STAHL-ELECTRONICS by using MA46H202 Macom a varactor diode.



Figure 4.19: Variation of resonance frequency, quality factor and signal-to-noise ratio with the change in biasing voltage of the varactor diode. The measurements shown here are done with the varactor heater set of 2 V and heating power of 200μ W, which is expected to raise the temperature of the varactor board.

for cyclotron resonator and not for the axial resonators.

4.5 The 65 GHz microwave system

In order to perform laser-microwave double-resonance spectroscopy on highly charged ions, the ions are confined in a Penning trap. The magnetic field leads to the Zeeman splitting of the levels which can be probed using tunable microwave radiation. In case of Ar^{13+} , the Zeeman transitions in $2P_{1/2}$ corresponds to a frequency of 65 GHz, whereas in $2P_{3/2}$ corresponds to 130 GHz. The microwave system for 65 GHz has already been developed and installed at ARTEMIS [103].

The microwave source, GT9000 Microwave Synthesizer from GIGATRONICS, is capable of generating microwaves from 2 GHz to 20 GHz with a power range of -20 dBm to 20 dBm. An external frequency standard rubidium clock FS725 from STANFORD



Figure 4.20: Obtained microwave power from the CERNEX CFM1616X410-01 frequency multiplier with respect to the power from microwave generator at 16.25 GHz.

RESARCH SYSTEM is connected to the microwave generator in order to have stable frequency output. For the 65 GHz microwave system, the microwave generator is operated at a frequency between 16 GHz to 16.5 GHz such that the value after frequency quadrupler is between 64 GHz to 66 GHz. The output of the microwave generator is split into two, one of which is connected to EIP 578 Source Locking Microwave Counter to check the power and frequency stability of the microwave source. The other is fed into the frequency multiplier (X 4) CFM1616X410-01 form CERNEX using a low loss coax cable from SHF, SC-119/50-SB-B . As the frequency multiplier gets heated up considerably faster, hence a fan is connected to it. The supply to the quadrupler and fan is provided from the HP DC power supply capable of providing 0-20 V/0-3 A using a common BNC connector. The input at the quadrupler has a SMA connector, whereas the output is WR-12 waveguide terminal. The output power from the quadrupler can be further amplified by using power amplifier SP654-15-24W from SPACEK which has a listed output power of 23.5 dBm and a gain of 19.8 dB at a frequency of 65 GHz.

Based on the preliminary tests in order to check the output power from the frequency multiplier, it was observed that the frequency multiplier CERNEX CFM1616X410-01 was out of order. A graph representing the output power from the frequency multiplier at 65 GHz with respect to the input power to the multiplier from the microwave generator at 16.25 GHz is shown in figure 4.20, for two different measurement terms: 2019 and 2020. In the graph, the output power from the multiplier decreases to almost zero in 2020, indicating a defect.

The device is replaced with MI-WAVE 934EF-20/387 frequency multiplier (\times 4). The specifications for the MI-WAVE are given in table 4.4. The DC power supply

4. The ARTEMIS EXPERIMENT

should be carefully connected to the multiplier since a reverse biasing will destroy the multiplier.

Using the new frequency multiplier, the output power is sufficient enough so the amplifier would not be required anymore. In order to have a flexibility in the amount of power radiated, a mechanical attenuator 520-E from MI-WAVE operable in a frequency range of 60-90 GHz and a power range of 0-25 dB is included in the set-up. Both the input and output terminals of the power attenuator are WR-12 waveguide connectors, whereas at the vacuum flange of the experiment we have a SMA (1.85) connector. Thus, a waveguide to coax converter followed by a short coaxial cable is connected to facilitate this connection.

For this microwave assembly, ARTEMIS uses a hybrid design that is a combination of WR-12 waveguide and a coaxial cable for the transmission of microwaves. Various aspects were taken into consideration while designing the 65 GHz microwave assembly:

1. Low transmission loss

The microwave source is positioned outside the trap bore which leads to a significantly long distance between the source and the ions (about 1.5 m). Thus, it is necessary that the transmission losses are minimum. The coaxial cable have higher losses due to the dielectric isolation. For this reason, the rectangular waveguides have an advantage over the coaxial cables.

2. Non-magnetic materials

ARTEMIS has a magnetic field of 7 T and even at the top of the magnet bore, there is a field of a few Gauss. The whole assembly for microwave transmission is in a region of strong magnetic field. The materials should thus be chosen in such a way that they do not disturb the magnetic field and can sufficiently transmit in the conditions of the experiment.

3. Cryogenic temperatures

The second stage in ARTEMIS is at ~ 4 K. Since the source of microwave is at room temperature, the transmission lines should be able to transmit efficiently at the cryogenic temperature alongside having least transmission of heat and low heat load. The rectangular waveguides have large surface area and have higher heat conduction in comparison to a coaxial cable.

4. High vacuum conditions

The microwave assembly is present in the magnet bore where the pressure is roughly 10^{-9} mbar. In order to maintain a good vacuum pressure, such a vacuum

15.0 to $22.5\mathrm{GHz}$
60 to 90 GHz
$5.0 \mathrm{dBm} (10.0 \mathrm{dBm} \mathrm{max.})$
21.0 to $23.9\mathrm{dBm}$
SMA-F (in), WR-12 Waveguide (out)
$6\mathrm{Vdc}$
$500\mathrm{mA}$

Table 4.4: Specifications of the 934EF-20/387 frequency multiplier (×4) from MI-WAVE.

feedthrough at the flange should be commercially available and the transmission lines should be vacuum compatible in this pressure range.

5. Space restrictions

Due to the space restrictions in the magnet bore, a flexible transmission line such as a coaxial cable is preferred over the rigid waveguide as it eases the set-up.

The waveguide installed in this hybrid design is made from OFHC copper and is bent at a small angle about 5 cm from the trap end, where it connects to the horn antenna pointing towards the trap. The design of the microwave system including the inside and outside assembly is shown in figure 4.21.

In order to test the components and transmission outside the ARTEMIS magnet bore, a power diode 950E from MI-WAVE is used. The output from the power diode is in terms of voltage which can be related to the incoming microwave power by the video sensitivity of the device. The device can be operated in a frequency range of 60-90 GHz and has a video sensitivity of 500 mV/mW. It connects to a WR-12 connector at the input and an SMA at the output.

A theoretical estimate of the losses in the above set-up comes out to be roughly 13.2 dB and a detailed description of this estimation is given in [103]. Additionally, the experimental measurement of the power loss is calculated to be 12.9 dB, which matches with the theoretically estimated value. Further loss of about 24 dB is expected from the materials between the horn antenna and the ions, major contributions coming from the ITO window (section 4.2.3), accounting for a total loss of 37 dB. Thus, the ion cloud in ST is irradiated with an approximate power between -61 dBm to -12 dBm (or 0 to 50 μ W).

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Figure 4.21: Schematic design of the 65 GHz microwave system implemented at ARTEMIS.

4.6 Optical detection system

For the double-resonance spectroscopy of Ar^{13+} at ARTEMIS, the fine structure transition is probed using a frequency stabilised laser with a wavelength of 441 nm. The laser can be tuned over several gigahertz with a sub-megahertz resolution. A secondary laser system is used to achieve the stabilisation. The optical detection system developed by A. Martin [112] consists of a 441 nm spectroscopy laser, a 452.756 nm master laser, a transfer cavity and a Doppler-free tellurium spectroscopy.

Figure 4.22 shows the schematic overview of the laser system along with the optical detection system. The laser set-up is positioned in the laser lab, which is located



Figure 4.22: Schematic design of the 441 nm laser system for Ar^{13+} at ARTEMIS [43]. The laser is transmitted through the 27 m long optical fibre to the vacuum feedthrough at the spectroscopy flange on top of the superconducting magnet and into the trap through another 2 m long optical fibre. The fluorescence light is transmitted via the image guide and focused on the camera or CPM.

beneath the HITRAP platform. As mentioned above, the laser system consists of two lasers: a) a custom-made external cavity diode master laser (ECDL) with a wavelength of 452.756 nm, built and characterised by A. Martin [112] and P. Baus [113] and b) a commercial ECDL TOPTICA DL 100 pro laser with a tunable wavelength between 439.4 nm to 445.8 nm and a maximum output power of 16 mW. For the purpose of frequency stabilisation of the spectroscopy laser to the master laser, a transfer cavity is installed which acts as a resonator. It consists of two silver-coatedplano concave mirrors which have a reflectivity of 93% at a wavelength of 441 nm. The master laser is the heart of the stabilization scheme. It is frequency-stabilised to the tellurium spectroscopy and is used to keep the free spectral range of the transfer cavity constant. Since there are various optical transitions of tellurium that are also well documented (tellurium atlas [114]), it is used as an absolute frequency reference for the spectroscopy laser and for the frequency stabilisation of the master laser.

Using a polarizing beam splitter (PBS), the light from the master laser is divided into two beams. One of these beams is overlapped with the spectroscopy light for the tellurium spectroscopy and the other is coupled into the transfer cavity. After heating the tellurium cell to a temperature of around 500 K, Doppler-free Lamb-dips



Figure 4.23: Photograph of the laser optics mounted at the four-leg structure in ARTEMIS. The fibre is mounted on a thin copper strip (fibre holder) and is pointed towards the trap. The horn antenna transmits the microwave radiation for double-resonance spectroscopy.

are observed by overlapping a probe beam with a counter-propagating saturation beam [115]. The master laser is locked to one of the Lamb dips to stabilise the frequency. By locking the length of the transfer cavity to the frequency of the master laser, the spectroscopy laser is locked to the master laser. In order to achieve this locking, the light of the spectroscopy laser is frequency shifted by the double-pass acousto-optic-modulator (AOM) and is locked to a different mode of the transfer cavity. Finally, the laser beam is set to a proper frequency and is transmitted to the experiment through a 27 m long optical fibre.

The laser light transmitted through the optical cable is coupled to the vacuum feedthrough on the spectroscopy flange. On the vacuum side, this feedthrough is connected to a 2 m long optical fibre whose insulation is removed at end pointing towards the trap chamber. This bare end is fixed to a thin copper strip and the light is collimated and focused using two aspheric lenses (figure 4.23). The ion cloud confined in ST can be illuminated by a laser with a beam diameter of roughly 3 mm.

The trapped ions in the spectroscopy trap would get excited after shining the laser beam and emit fluorescence radiation. This fluorescence radiation passes through the ITO window and gets collimated by the same two aspheric lenses mentioned above. The collimated light is focused onto the 'image guide' which consists of a fibre bundle structured in a 72×72 array with an area of 16 mm^2 . After transmission through the image guide, it is out-coupled from the magnet bore through a vacuum window at the flange. This light can be detected using *a*) ImagEM X2 Em-CCD camera from HAMAMATSU or *b*) a channel photon multiplier MP-984 CPM from Excelitas. In order to purify the observed fluorescence light, a band pass filter with a bandwidth of 10 nm around the center wavelength of 441 nm and a line filter with a bandwidth of 1.7 nm also at the center wavelength of 441 nm are implemented in the beam path along with two extra collimator lenses. A shutter is installed in the laser beam path outside the magnet bore in order to manage the timings of the laser illumination of the ions and of the fluorescence detection. This protects the detectors from over-exposure.

The laser light produced through this system can thus be used along with the microwave radiation (section 4.5) in order to implement laser-microwave double-resonance spectroscopy on the ion cloud confined in the spectroscopy trap.

Chapter 5

Investigation of ion ensembles and trap characterisation at ARTEMIS

The ARTEMIS experiment aims to undergo high-precision measurement of the qfactor of electrons bound to heavy, highly charged ions using laser-microwave doubleresonance spectroscopy technique (section 3.3). To reach this milestone, a sufficiently cooled ensemble of ions with a single ion species needs to be stored in the spectroscopy trap (ST, section 4.2.2). In this work, investigations are performed on boron-like argon (Ar^{13+}) ions to study the behavior of the ion cloud in both the creation trap and the spectroscopy trap. A detailed discussion concerning the creation of highly charged ions, ion ensemble cooling using successful transport between the two traps, comparison of selective cleaning of the cloud to obtain a single ion species in both of the traps and estimation of pressure is presented in this chapter. The knowledge of the trap capacitance is essential to the experiment, all the more so in the development of non-destructive detection systems. The design parameters of these resonators (section 4.4) are influenced greatly by the load capacitance. Therefore, the capacitance values of both the traps have been estimated in order to analyse their effect on the properties of non-destructive detection system, such as resonance frequency, quality factor and signal-to-noise ratio (SNR).

The results described in this chapter regarding ion cloud investigation are based on the data acquired during the 2019 and 2021 measurement campaigns. Measurements concerning the estimation of trap capacitance and its influence on the detection system have been carried out in 2020. Unless otherwise stated, the data is acquired using the KEYSIGHT N9000B-CXA spectrum analyser which is controlled by the LABVIEW measurement system. The results presented in this project along with the discussions in [34, 43, 103] provide a complete understanding of the ion ensemble properties.

5.1 Estimation and characterisation of trap capacitance

The resonators connected to ARTEMIS act as non-destructive detectors and also help in cooling the ion cloud. This is possible only when the detection circuit has a frequency equal to the frequency of motion of ions (section 4.4). These resonators have a frequency determined by the self capacitance and inductance of the respective coils. In addition to this, the resonance frequency is affected by the capacitance of the trap which acts as a load capacitance. The term 'trap capacitance' refers to the capacitance of the system due to the trap electrodes, wires, trap can and all other components (except the self capacitance of the inductor coil) which contribute to the load capacitance of the system to the electronic detectors at their point of connection. As seen in table 5.4, the center frequency of the resonators change significantly on connection to the trap. Thus, it becomes important to characterise and measure the trap capacitance for various cases, such as on attaching the super-insulation foil, the 40 K radiation shield, after inserting the trap in magnet bore and most importantly after cooling at 4 K. These measurements are performed after excitation with the tracking generator of the KEYSIGHT spectrum analyser with a power of -50 dB m and have been explained in detail in this section. Throughout these measurements, the experimental set-up is kept the same (with room-temperature filter boards, connectors and spectrum analyser) as used during measurements with trapped ions.

In the ARTEMIS set-up, three resonators have been connected to the Penning trap: ARES CT resonator connected to E13, ARES ST connected to E2 and CRES ST attached to E4 electrode. Characteristics and geometry of these three coils have been given in table 4.2 in section 4.4.

The 'ambient conditions' listed in this section correspond to:

- Resonator at test-stand: Detection circuit (resonator along with cryogenic amplifier) tested outside the ARTEMIS set-up at the test-stand (appendix A) at room temperature and atmospheric pressure.
- Resonator connected to trap: Detection system mounted at the four-leg structure of the trap with the hot-end connected to the signal line from the trap.
- Super-insulation foil attached: In order to avoid radiative heating and reduce the heat load on 4 K stage, a super-insulation foil covers the four-leg structure.
- Radiation shield attached: The 40 K radiation shield is attached to the set-up.

- In magnet and evacuated: The set-up is put into the magnet bore with a magnetic field of 7 T and is evacuated to 10⁻⁸ mbar using a turbo molecular pump. The measurement in this configuration of the set-up becomes more important in the case of CRES ST. This is due to the presence of varactor diode which gets affected by the strong magnetic field [103].
- Cooled to 4 K: The apparatus is cooled down to cryogenic temperature such that the detection circuit is operating at ~ 4 K, 7 T magnetic field and a pressure of better than 10^{-9} mbar in the magnet bore.

Tables 5.1, 5.2 and 5.3 describe the effect of the above listed conditions on the resonance frequency, quality-factor and signal-to-noise ratio (SNR) of the detection circuits. The load (system) capacitance value is estimated for each of the cases and is depicted in table 5.4.

Table 5.1: Frequency measurements under various trap circumstances. The table shows the values of the resonator frequency as measured using the spectrum analyser.

	Frequency [MHz]			
Ambient conditions	ARES CT	ARES ST	CRES ST	
Resonator at test-stand	1.0361	0.8930	102.34	
Resonator connected to trap	0.6964	0.7493	32.804	
Super-insulation foil attached	0.6992	0.7444	33.026	
Radiation shield attached	0.6959	0.7473	33.232	
In magnet and evacuated	0.6959	0.7474	32.826	
Cooled to 4 K	0.7058	$0.74\overline{67}$	37.112	

Table 5.1 shows the changes in the resonance frequency at different conditions. Upon connection to the trap, the resonance frequency changes significantly. This is because the trap capacitance is in parallel with the self capacitance of the resonance coil and, according to equation $f = 1/2\pi\sqrt{L(C_p + C_L)}$, it changes the central frequency of the detection circuit. There are significant changes in resonance frequency upon cooling the trap to 4 K. The shift in the frequency is notable for the cyclotron resonator. This is because the system contributing to frequency at E4 also consists of a varactor diode. The capacitance of the varactor diode is comparable to the self capacitance of the CRES coil, thus leading to the large frequency shift. Additionally, the effect of cooling is also more pronounced for the CRES ST. This can again be attributed to the varactor diode, whose properties change significantly in the combination of strong magnetic field and low temperatures [43]. The value of CRES ST

	Q-factor			
Ambient conditions	ARES CT	ARES ST	CRES ST	
Resonator at test-stand	50	50	116	
Resonator connected to trap	33.4	40.1	68.1	
Super-insulation foil attached	34.5	41.7	59.9	
Radiation shield attached	34.6	42	64.5	
In magnet and evacuated	34.1	40.9	52.9	
Cooled to 4 K	376.1	345.5	33.5	

Table 5.2: Variation of the Q-factor of the detection circuit with different load capacitance from the

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system.

frequency, SNR and quality factor at 4 K listed here are measured without heating the versator diadal. The versition of CRES ST properties with versator diada bias

the varactor diode¹. The variation of CRES ST properties with varactor diode bias potential is depicted in figure 4.19 and in appendix B.

The effect of the different load capacitances on the Q-factor and SNR of the three detection systems connected at ARTEMIS is shown in tables 5.2 and 5.3. As discussed above, the shifts are more prominent in CRES ST because of the presence of varactor diode. When the detector circuits are cooled to cryogenic temperature, the quality factor goes up multiple folds, except for CRES, where the varactor diode quality restricts the Q-factor of the system. It can be seen from figure 4.19, that on raising the diode temperature, and changing the bias potentials, the Q-factor varies by a factor of ~ 2 .

	Signal-to-noise ratio [dB]			
Ambient conditions	ARES CT	ARES ST	CRES ST	
Resonator connected to trap	30.5	31.7	32.3	
Super-insulation foil attached	29.5	30.9	31.2	
Radiation shield attached	29.1	30.9	31.9	
In magnet and evacuated	28.9	32.7	36.9	
Cooled to 4 K	56.7	57.3	32.5	

Table 5.3: Variation of signal-to-noise ratio of the detection circuit with different load capacitance from the system.

The measurement for the coil characteristics (frequency, quality factor, SNR) are

¹The varactor diode introduces a higher capacitance at 4 K and 7 T field and is heated to have optimum operational frequency and higher quality factor. More details have been discussed in section 4.4.3.

performed at the test-stand (Appendix A) outside the ARTEMIS experimental setup. In order to measure the self- capacitance of the coil, various high quality factor capacitors from JOHANSONS have been connected in parallel to the hot-end of the coil (figure 4.8). From the frequency values listed in table 5.1, and the self capacitance of the coil, the capacitance of the system under different configurations is depicted in table 5.4.

Table 5.4: Estimation of the capacitance value for different settings of the system for the three resonators connected to different trap electrodes. The values are estimated using the frequency shifts in the non-destructive detection circuit.

	Capacitance [pF]			
Ambient conditions	ARES CT	ARES ST	CRES ST	
Resonator connected to trap	39.5	11.5	35.1	
Super-insulation foil attached	38.9	11.7	34.6	
Radiation shield attached	39.6	11.4	34.1	
In magnet and evacuated	39.6	11.4	35.0	
Cooled to 4 K	37.6	11.2	26.5	

The knowledge of the trap capacitance values is crucial in designing the resonator coil suited to a particular resonance frequency. The measurement process was repeated two more times (each time when the trap was taken out of the magnet bore), and the values of the load capacitance were found to be consistent within $\pm 5\%$.

5.2 Ion creation

After connection to the HITRAP beamline, ARTEMIS will be able to receive ions from the external ion sources such as EBITs and highly charged ions available from the GSI facility. Until that milestone is achieved, the creation trap of ARTEMIS acts as a mini-EBIT and ions are created inside the Penning trap using the field emission point (FEP, section 4.3).

As a brief overview of the creation process described in section 4.3: for the production of ions inside the ARTEMIS trap, the first step is to apply alternating potentials on the trap electrodes in CT (± 250 V) so as to form three consecutive harmonic traps [43]. The FEP electron gun is then set up by applying high negative potentials to the tungsten tip (\sim -1100 V to -1600 V) and a relative high positive potential on the accelerator electrode. The reflector electrodes are set at least 250 V lower than the FEP voltage to trap the electrons for charge breeding (figure 5.2). Argon gas is injected



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Figure 5.1: Schematic of the *in-situ* creation of ions using electron impact ionisation in the ARTEMIS creation trap.

into the trap, by using either one or both of the heaters attached to the cryogenic diffusion valve. The flow rate of the argon gas injection can be controlled through the dosing valves. Thus, argon ions are created and confined in the three traps of CT. Figure 5.1 shows the schematic of the ion creation process. By modifying the potentials on the trap electrodes, the ions are merged to a single trap E 13. There are several free parameters which can be varied to optimise the ion creation. Since the creation parameters differ after every thermal cycling of the trap, the recalibration (fine-tuning) should be performed before every measurement campaign. The problem of optimisation boils down to finding the best set of values for a) FEP electron gun operation and b) gas injection system.

FEP electron gun operation

A high negative voltage applied to the field emission point enables the electrons to tunnel out of the tungsten tip. The energy of the emitted electrons depends strongly on the applied FEP voltage while the current density depends on the accelerator voltage and shape of the tip. The reflector electrodes are set to high negative potentials which traps the electrons over the length of CT. The electrons are reflected backand-forth for a breeding time t_c , leading to the creation of high charge states. Thus, an optimum set of values for a) FEP voltage (V_{FEP}), b) accelerator voltage (V_{acc}), c) reflector voltage (V_{acc}) and d) breeding time (t_c) has to be determined.



Figure 5.2: Variation of current on the ITO window as a function of voltage on the reflector electrodes for different values of V_{FEP} . For low values of reflector voltages, the electrons can fly all the way to the ITO (section 4.2.3), which acts as a Faraday cup and the current is measured from the ammeter connected to E 1. It can be observed from the graph, that current on ITO is zero when the reflector voltage is at least 250 V lower than FEP voltage.

Figure 5.2 shows that the value of reflector voltage should be at least 250 V lower than the FEP voltage, in order to confine all the emitted electrons in CT. During the measurement campaigns of 2019 and 2021, the values shown in table 5.5 are observed to be favorable towards creation of the desired Ar^{13+} charge state.

It has been observed that the current in the beginning of the first few creation attempts is usually lower than during the continued operation of the FEP during the measurement campaign. This effect can be attributed to the build-up of ice on the tip which can be substantially removed by application of high voltage on the tip for short time.



Figure 5.3: Comparison of FEP currents as a function of voltage on the accelerator electrodes for different FEP voltages in 2019 and 2021. The current varies in each measurement campaign, hence there is a need of optimisation of the creation parameters after every thermal cycling.

Gas injection system

Neutral argon gas is injected into the trap through the gas injection system described in section 4.3. The optimisation of the gas injection parameters is crucial for ion creation. In order to insert the gas, the cryogenic gas valve is heated and then the pressurised air-valve is opened for a short period of time. The amount of gas inserted into the trap depends on a) the duration for which cryogenic valve is heated (t_{heater}) , determining the final temperature attained, b) gas flow time (t_{gas}) , which is the time for which pressurised air valve is open, and c) pressure of the gas in the injection system (P_{gas}) which is controlled by using the dosing valves.

It has been observed that the injection parameters are different for the measurements done in 2019 and 2021. The major difference in the heating time is due to a short circuit on one of the heaters connected to the cryogenic valve. This was rectified before the measurements performed in 2021, leading to shortened heating times. The set of parameters listed in table 5.5 ensured dominant creation of Ar^{13+} in the trap. However, it should be noted that the combination of values given in table 5.5 is not the only possible optimum set. Due to the large number of variables, it is possible to have other combinations which largely create the desired Ar^{13+} ions.

Table 5.5: Optimised parameters for the efficient creation of Ar^{13+} during measurements performed in 2019 and 2021.

	$V_{\rm FEP}$	$V_{\rm acc}$	$V_{\rm ref}$	t_c	$t_{\rm heater}$	$t_{\rm gas}$	$P_{\rm gas}$
2019	-1800 V	$2200\mathrm{V}$	$-2200\mathrm{V}$	$>40\mathrm{s}$	18 sec	$> 48\mathrm{ms}$	$1\mathrm{mbar}$
2021	$-1600\mathrm{V}$	$1650\mathrm{V}$	$-2000\mathrm{V}$	${\sim}36\mathrm{s}$	8 sec	$>\!\!64\mathrm{ms}$	$1.3\mathrm{mbar}$

5.3 Mass spectrum read-out

The mass spectrum or q/m spectrum is an integral measurement technique used to resolve ion species confined in the ARTEMIS Penning traps. The spectrum is obtained by sweeping the trapping voltage for a fixed resonance detection frequency and a plot of the ion signal versus trap voltage is obtained. The axial frequency is related to the applied electric potential on the trap as $\nu_z = \sqrt{QC_2U_0/4\pi^2Md^2}$ (section 2.2) such that, for a particular frequency of the RLC circuit, ions with different charge-to-mass ratios come into resonance at different trapping voltages. This resolves the ions with different charge-to-mass ratios. Such a spectrum is obtained using the 'zero-span' mode of the spectrum analyser N9000B-CXA from KEYSIGHT TECHNOLOGY.



Figure 5.4: Mass spectrum obtained by sweeping the trapping potential in the creation trap of ARTEMIS. Different charge states of argon are resolved and come into resonance with the center frequency 706.632 kHz of the ARES CT resonator. δU_0 is the full-width half-maximum (FWHM) corresponding to broadening of the ion signal and ΔU_0 is the shift in peak position relative to the theoretically estimated value.

In the 'zero-span' mode, the local oscillator of the spectrum analyser does not sweep but is rather fixed to a frequency (in this case, the resonance frequency of the RLC circuit) with a narrow bandwidth. The value at every voltage step in the mass spectrum is equal to the signal amplitude integrated in this bandwidth. Hence, the peak represents the height of the ion signal on top of the resonator signal (as seen on the spectrum analyser in the frequency space) and the baseline of mass spectrum corresponds to the signal amplitude at the central frequency of the resonator.

The q/m spectrum is a powerful tool to characterise the ion cloud based on its temperature, number density, plasma parameter and so on. As different ion species come into resonance with the RLC circuit at different trapping voltages, it also leads to resistive cooling of the ions. Some properties of the spectrum that can be used to characterise the ion ensemble are:

- Signal height: The amplitude of the signal is related to the amplitude of the ion motion. As the ions cool, the signal amplitude reduces until the ions are in thermal equilibrium with the resonator.
- Area under the peak: The area under the peak in a mass spectrum is a convolution of kinetic energy of a single particle and the total number of ions in

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that charge state. For the ion clouds stored in ARTEMIS, the number of ions in a charge state does not change significantly between two consecutive ramps due to long storage lifetimes and negligible collisions. Hence, the area under the signal peak is a measure of individual particle energies.

Shift in peak position: As depicted in figure 5.4, ΔU₀ shows the shift of the peak from the expected theoretical voltage of the peak. This shift can primarily be attributed to the space-charge effects and field imperfections (sections 2.3.2, 2.3.3). The contribution of the space charge effect towards the shift in the axial frequency is given by:

$$\nu_z' = \nu_z \sqrt{1 - \frac{\nu_p^2}{3\nu_z^2}} \qquad \text{with} \qquad \nu_p^2 = \frac{nq^2}{4\pi^2\epsilon_0 m} \tag{5.1}$$

where n is the ion number density and ν_p is the plasma frequency. The frequency shift can be obtained from the q/m spectra in order to estimate the number density of the trapped ion ensemble.

• Peak width: The width of the signal peaks is an important parameter towards the measurement of temperature of the ion cloud. The energy of a thermalised ion cloud at a temperature T follows a Boltzmann distribution. Due to significant electric field imperfections, the ion frequency depends on its energy. Hence, for an ion cloud with a thermal distribution, there is a corresponding distribution of its motional frequency. This leads to shifting and broadening of the signal peaks. The temperature T of the ion cloud can hence be determined from the relative width $\delta \nu_z / \nu_z$ of frequency distribution such that:

$$\frac{\delta\nu_z}{\nu_z} \approx \frac{3C_4}{C_2^2} \frac{k_B T}{qU_0} + \frac{15C_6}{C_2^3} \left(\frac{k_B T}{qU_0}\right)^2 \tag{5.2}$$

where k_B is the Boltzmann constant and C_2, C_4, C_6 are the known coefficients for a given trap. It should be noted that the quantity that can be obtained from the q/m spectrum is the width (FWHM) of the signal peak, δU_0 , which corresponds to width of the frequency distribution as:

$$\delta U_0 = 2\pi \sqrt{\frac{4md^2 U_0}{qC_2}} \delta \nu_z \tag{5.3}$$

Figure 5.4 shows a q/m spectrum for the voltage sweep in ARTEMIS creation trap using the axial resonator with a center frequency of 706.632 kHz. The grid lines in
the background represent the theoretically expected position for a single ion signal in the absence of any field imperfections. The signal peaks correspond to the ions with different charge-to-mass ratios. The ion peaks are shifted and broadened mainly due to the field imperfections and space charge effects. Hence, using the shift (ΔU_0) and the broadening (δU_0) values from the acquired spectrum and equations 5.1 and 5.2, an estimate on the ion number density and temperature of the cloud can be obtained.

5.4 Ion ensemble cooling and transport between the traps

In order to prepare the ion ensemble for double-resonance spectroscopy, the first step is to cool the ions confined in the ARTEMIS Penning trap. The significance and theory of ion cooling have been detailed in section 2.5. Among the various cooling techniques described in the section 2.5, a combination of resistive cooling and the cooling caused by the removal of hotter ions from the trap has been implemented in ARTEMIS. For the cooling of ions using this procedure, the presence of trap electrodes of different diameters play an important role. The concept of this combined cooling technique is explained in this section.

The ion transport is carried out slowly by changing the potential of one electrode at each time step. Figure 5.5 shows the schematic for a single step of transport procedure from electrode E14 to E13. The potential of the neighboring electrode E13 is lowered to form a wide potential well for the ions. Next, the potential at E14 is lifted slowly and the ions are finally trapped in E13. Since the switching time from the filterboards is at least a few milliseconds, the transport is always slow compared to the ion oscillation frequency of hundreds of kHz. The same process is used consecutively until the ions are trapped in E3, the ring electrode of the spectroscopy trap.



Figure 5.5: Schematic of the adiabatic ion transport from E 14 to E 13. The transport between the traps is performed by changing the potential of only one electrode at each time step, until the ion cloud is confined in E 3.

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Figure 5.6: Ion motion cooling as a result of transport between the two traps. The scan in red in (a) shows the first scan after ion production in the creation trap. The ion signal is blurred with no distinct ion peaks. The hot cloud from the creation trap is transported to the spectroscopy trap (red). After multiple transports ion ensemble gets cooled and discrete peaks are observed.

The high voltage electrodes E8 and E9 have smaller inner diameter (10 mm) than the diameter of electrodes in CT and ST (17.54 mm). The difference in the dimensions is crucial to the process of cooling by repeated transport. In a hot cloud of ions in ARTEMIS, it is fair to assume that the ion cloud almost fills the entire radial dimension of the trap. When a hot cloud of ions is transported from the creation trap to the spectroscopy trap, the ions with higher energy are stripped from the cloud and are no longer confined. This is because hotter ions are present towards the periphery of the ion cloud and are scraped off during transport through E8 and E9. Since the higher energy ions are kicked out of the trap, the remaining ions come into a

thermal equilibrium at a lower value (similar to evaporative cooling). The number of ions in the ensemble after each transport is reduced, and the remaining ions have less energy and are thus cooled. The process can be repeated multiple times in order to sufficiently cool the ions.

Figure 5.6 shows the cooling of the ion motions as a result of transport between the traps. The red curve in the creation trap shows the mass spectrum just after creation. The cloud is then adiabatically transported to the spectroscopy trap and a q/m scan is performed (red curve in spectroscopy trap). After a successful transport to the creation trap, the spectrum depicted in blue in figure 5.6a is observed. In each transport, the ion cloud gets colder and smaller. This is also depicted in figure 5.7. The evolution of area under the curve and the relative shift of the ion frequency for Ar^{15+} in both CT and ST is plotted as a function of number of transports. It can be seen that the area under the curve reduces after each transport. As explained in section 5.3, the area under the q/m spectrum is a convolution of ion number and the energy of ions. Since the pressure inside the trap is lower than 10^{-14} mbar, hence, the magnetron expansion of the cloud can be considered to be negligible. Hence, a decrease in area as well as the frequency shift relative to the reference frequency² indicates cooling of the ensemble. It should be noted that in each of these scans, the induced image currents also undergo power dissipation through the resonator circuit, thereby resistively cooling the ions.



Figure 5.7: Variation of the area under the curve and relative frequency shifts as a function of number of transports between the creation trap and the spectroscopy trap.

When the ion ensemble is sufficiently cooled, the area under the curve does not change significantly between two consecutive transports, indicating minimal loss in

 $^{^{2}}$ Resonance frequency for an ion in the ideal trap without space charge effects and field anharmonicities is considered as the reference frequency.

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the ion number. This can be seen in figure 5.8, where the transport of a cooled cloud of Ar^{13+} is shown from spectroscopy trap (red in (a)) to creation trap (red in (b)) and back to the spectroscopy trap (blue in (a)). The area difference between the scans before and after transport in ST is almost zero, indicating an efficient and successful transport procedure.



Figure 5.8: Transport of a cooled ion cloud between the two traps at ARTEMIS. A cooled cloud of Ar^{13+} ions from ST (details on cloud cleaning in section 5.6) is transported to CT, depicted by red curve in (b). The ion cloud is then transported back to ST with minimal loss in ion number. The red curve in (a) is the mass scan before the transport and blue scan is measured after the transport back to ST.

Therefore, cooling by multiple transport between the traps can be utilized as a cooling technique only for hot clouds and is even faster than the resistive cooling. The application of resistive cooling on the ion ensemble stored in ARTEMIS has been discussed in detail in the previous reports by M. Wiesel [103] and S. Ebrahimi [43] and depicted a cooling time of a few days. Similar temperatures can be achieved in a couple of hours by implementing the transport procedure. Hence, the transport cooling process is faster than the resistive cooling, and can be implemented easily in the trap by varying the trap potential. It is specifically effective for a hot cloud of ions and was implemented routinely together with resistive cooling for better efficiency, during the measurement campaigns of 2019 and 2021.

5.5 Magnetron cooling

As defined in section 2.2, the magnetron motion of confined ions has negative energy states. Hence, the generic concept of cooling to decrease the energy of motion does not apply to magnetron motion. A decrease in energy of the magnetron state pushes it down in the energy ladder, leading to an increase in the motional amplitude. In order to cool the magnetron motion, it has to be coupled to one of other two oscillations, i.e. axial or modified cyclotron motion and cooling the second motion. Since the axial motion of the ions in CT can be cooled by using the RLC circuit, axial sideband coupling is performed through one of the segments of electrode E 15 (section 4.2.1). When the ions are irradiated with $\nu_z + \nu_-$, both axial and magnetron motional states climb up the energy ladder. This heats the axial motion, thereby increasing its amplitude of oscillation. However for the magnetron motion, going up in energy causes cooling of the motion and hence reduces its amplitude. The ions are thus magnetron cooled. To cool the axial motion along with the magnetron cooling, the trap depth is set to a trap potential of the corresponding charge state. This brings the ions in resonance with the axial resonator and resistively cools the axial motion. This is referred to as magnetron centering [50]. An important aspect to note is that for an ion ensemble with different charge-to-mass ratios, only a single ion species can be magnetron centered.



Figure 5.9: (a) Energy ladder depicting the quantised states of ion motion and the process of magnetron centering. The irradiation of ions with $\nu_z + \nu_-$ while resistively cooling the axial motion results in the magnetron centering of the charge state. (b) Magnetron centering for Ar^{10+} ions confined in CT of ARTEMIS. The change in ion peak width before and after irradiation indicates magnetron centering.

Figure 5.9 shows the magnetron cooling for Ar^{10+} charged state. In ARTEMIS, ions experience a magnetic field of 7 T. When an axial resonator of 703.5 kHz is attached to the creation trap, Ar^{10+} ions come into resonance with this at a trap voltage of 65.4 V. Using equation $\nu_{-} = \nu_c/2 - \sqrt{\nu_c^2/4 - \nu_z^2/2}$ defined in section 2.2, the magnetron frequency for Ar^{10+} is calculated to be 9.2 kHz. A signal with the frequency range of 708.6 kHz to 725.9 kHz having 100 bursts, 100 ms excitation time and $V_{pp} = 0.2 \text{ V}$ is irradiated on the ions with the trap voltage fixed at 65.4 V. A decrease in the width of Ar^{10+} ion peak indicates magnetron centering. However,

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Figure 5.10: Ion ensemble consisting of highly charged argon and tungsten ions confined in ARTEMIS creation trap. During ion creation, highly charged tungsten ions are also formed and stored in the trapping fields in ARTEMIS. Such a spectrum can be 'cleaned' using SWIFT to obtain a single charge state.



the same effect is not observed for Ar^{9+} peak since the irradiation and cooling was focused towards Ar^{10+} frequencies.

5.6 Ion charge state selection using SWIFT

The process of ion creation explained in section 4.3 (also in-flight capture) confines ions with different charge-to-mass ratios in the trap. Along with the desired argon ions, other 'impurities' such as nitrogen and oxygen ions and even highly charged tungsten ions are observed in the trap³ (figure 5.10). In order to perform the lasermicrowave double-resonance spectroscopy on Ar^{13+} ions, the ion cloud should be cooled and cleaned to have only the desired Ar^{13+} ion ensemble in the trap. This can be done by using the Stored Waveform Inverse Fourier Transform (SWIFT) technique. To remove the unwanted ions from the trap, their axial ion motion is excited by irradiating a radio-frequency signal and lowering the trap depth such that particles with higher energy are no longer confined. The rf signal consists of a frequency band of all unwanted particles. The trap is hence 'cleaned' to have the requisite charged particles. The method employs a fast Fourier transform to convert a signal from the time domain to the frequency domain.

In the past measurements for SWIFT reported in [43] and [103], this technique has been applied in CT to clean the cloud before transporting it to the ST in ARTEMIS. In this work, the application of SWIFT on the ions confined in ST is reported for the first time.

³Highly charged tungsten ions are observed to be produced in the ARTEMIS creation trap. The observation complements the tungsten ions observed in previous reports by M. Wiesel [103] and S. Ebrahimi [43]

5.6.1 In the creation trap

In order to selectively eliminate the ions from the creation trap of ARTEMIS, the SWIFT signal corresponding to the frequencies of unwanted ions is irradiated using the transmission line connected to one half of the split electrode E 13. The room temperature filter boards [43] limit the power that can be applied to a value of $10 V_{pp}$. As described in [116], the excitation voltages should be of the order of $100 V_{pp}$. Therefore, the entire frequency band corresponding to unnecessary ions is divided into multiple small bands and the irradiation is done by targeting a small fraction of ions at each step. Once the ions are excited to higher energies, the potential on the ring electrode E 14 is switched rapidly to facilitate ion extraction. The fast switching is done using the fast voltage switch from STAHL ELECTRONICS by keeping the switching time t_{switch} much shorter than the thermalisation time $(\tau_T)^4$. Thus, a large number of cycles of excitation and switches are repeated corresponding to different frequency bands with each axial SWIFT excitation comprising of 1000 bursts with $8 V_{pp}$ signal amplitude and a switching time of $600 \,\mu s$. During the irradiation, the trapping electrode E14 is at -250 V and the adjacent electrodes are at 10 V, giving a trap depth of 260 V. After each SWIFT cycle, at least one q/m scan is acquired to confirm the presence of desired ions and decide the SWIFT band for the next cycle. Furthermore, the irradiation of SWIFT signal may heat up the required ions, thereby necessitating resistive cooling between two SWIFT excitations.

Based on the theory given in [117], the SWIFT technique has been implemented in the LABVIEW control system of ARTEMIS by M. Kiffer [116]. More details on implementation of SWIFT in CT can also be found in the earlier works of M. Wiesel [103] and S. Ebrahimi [43].

Figure 5.11 shows the step-by-step procedure for obtaining a pure Ar^{10+} ion cloud in CT. A certain fraction of ion cloud has been irradiated at each step and the cloud is resistively cooled between two consecutive steps. Additionally, a comparative study between the cleaning of a cooled cloud vs a 'hot' cloud has been performed and depicted in figure 5.12. As can be seen from figure 5.12a, for a cloud which is not sufficiently cooled before application of SWIFT mechanism, the area under the peak for Ar^{13+} reduces to 25% of the peak area before SWIFT. In contrast to this, upon cleaning the ion ensemble for Ar^{10+} state in a cooled cloud, the area under the

⁴Thermalisation time of ion cloud in ARTEMIS is $\sim 6.7 \text{ ms}$. The trap depth should be decreased faster than the thermalisation time in order to avoid transfer of energy to the desirable ions, leading to their extraction as well.



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Figure 5.11: Charge state selection using SWIFT technique to obtain a pure Ar^{10+} ion cloud in CT. The SWIFT technique is implemented in multiple steps at ARTEMIS. The blue rectangles represent the voltage window over which the SWIFT excitation is done. After each SWIFT step, mass spectra are recorded in order to detect and cool the ions.

peak (hence the ion number) is 95% of its area before SWIFT. This indicates the effectiveness of SWIFT for charge state selection on a cooled cloud of ions.

5.6.2 In the spectroscopy trap

Ion cloud transported to spectroscopy trap can have a distribution of different charge states. Even if a 'cleaned' cloud from creation trap is transported to the spectroscopy



Figure 5.12: Implementation of SWIFT for charge state selection in (a) hot (b) cooled ion clouds in the creation trap. The mass spectrum in red represents the ion cloud signal before selectively cleaning the cloud, while blue spectrum represents the single charge states after applying multiple steps of SWIFT excitation.

trap, lower charge states can be observed in ST after a certain time.⁵ This is due to charge exchange with the residual gas in the trap. Therefore, the trapped ion ensemble now has multiple charge states, requiring further SWIFT. This issue can be approached in two different ways, a) by transporting the ions to CT, performing the cleaning and transporting it back to ST, or b) performing SWIFT on the ion ensemble in ST. Hence, application of the SWIFT technique on the ions stored in the spectroscopy trap was studied to examine the effectiveness and feasibility of the charge state selection in ST in comparison to SWIFT of ions in CT.

The LABVIEW program for SWIFT in creation trap calculates the frequency band to be applied for excitation from the input values of lower and upper limits of voltages. An extension of this SWIFT program to spectroscopy trap firstly converts the input voltage values indicated from the mass spectra in ST to the corresponding voltage band in CT. This is performed using the ratio of frequencies of the corresponding resonators, as indicated in equation 5.6. This equivalent voltage band is input for the existing SWIFT program with an exception of fast switching. In the charge state selection procedure in the spectroscopy trap, fast switching for the ring electrode is not implemented, in contrast to the creation trap where the potential on E 14 is switched to remove unwanted ions from the trap. The remaining input parameters are same as for SWIFT in CT, i.e. 1000 bursts with amplitude of $8 V_{pp}$ for an excitation time of 1 ms.

If U_i is the trapping potential depth at which ions with a particular q/m value come into resonance, the center frequency of the axial resonators connected to ARTEMIS

 $^{{}^{5}}$ For the confinement time comparable to the lifetime of ions.

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traps $(\nu_{res,i})$ is defined as:

$$\nu_{res,i} = \frac{1}{2\pi} \sqrt{\frac{qC_2 U_i}{md^2}} \qquad ; \qquad i \in [\text{CT, ST}].$$
(5.4)

The same equation can be used to derive the frequency band of SWIFT corresponding to the input voltage values. For the voltage band with $V_{i,j}$; $j \in (lower, upper)$ as the lower and upper bounds for SWIFT, the corresponding frequencies are:

$$\nu_{i,j} = \frac{1}{2\pi} \sqrt{\frac{qC_2 V_{i,j}}{md^2}} = \nu_{res,i} \sqrt{\frac{V_{i,j}}{U_i}}$$
(5.5)

Using the above two equations, the equivalent input voltage limits for the SWIFT programme from the ST voltage values derived from the mass spectrum in ST can be given as:

$$U_{\text{CT},j} = U_{\text{ST},j} \left(\frac{V_{CT,j}}{V_{ST,j}}\right) \left(\frac{\nu_{res,CT}}{\nu_{res,ST}}\right)^2$$
(5.6)

Thus, SWIFT signal obtained by providing these converted input values is irradiated on the ions through the transmission line connected to electrode E4 and hence excites the ions. These highly energetic ions after excitation are then lost, leaving the trap with a single ion species. It should be noted that the process of cleaning is done in several steps as in the case of SWIFT in CT.

The implementation of SWIFT technique is observed on hot and cooled cloud of ions and the area under the desired ion peak is compared. It has been observed that the area under the peak for Ar^{13+} in a hot cloud reduces to 14% of its value before SWIFT. However, SWIFT for Ar^{13+} ions in a cooled cloud brings the peak area down to 25% of its value before cleaning. This reinforces the efficiency of SWIFT application in a cooled cloud of ions.

From the above discussion, it can be seen that charge state selection can be applied successfully in both the traps at ARTEMIS: the creation trap as well as the spectroscopy trap. The effectiveness of application of charge state selection using SWIFT has been studied in both the traps using 'hot' and 'cooled' cloud of ions. In both the traps, selective cleaning was found to be more efficient in a cooled cloud of ions, as can be seen from figure 5.12 and 5.13. Additionally, through the comparison of the charge selection in a cooled cloud in creation trap and spectroscopy trap, it has been observed that the area under the peak for a cooled cloud in CT reduces only by 5%. However, the area reduces by 75% in comparison to the value before SWIFT in a cooled cloud in ST. The loss in area can be directly related to the loss in ion number for cooled ions. The greater loss in ion number in the spectroscopy can be



Figure 5.13: Implementation of SWIFT for charge state selection in (a) hot (b) cooled ion cloud in ST. The mass spectrum in red represents the ion cloud signal before selectively cleaning the cloud, while the blue spectrum represents the single charge states after applying multiple steps of SWIFT excitation.

attributed to the absence of fast switching of the ring electrode. Since the trap in ST is shallower as compared to the maximum achievable trap depth in the creation trap, it is possible to provide enough excitation to a selective range of ions and 'make' them escape. Although the ion cloud can be cleaned to achieve a single charged state, the lack of fast switching allows for thermalisation of the ions, thus allowing the desired ion species escape along with the unwanted ions. Therefore, it can be concluded that the SWIFT in creation trap with a cooled cloud of ions is the most efficient way of obtaining a cleaned cloud of ions.

5.7 Estimation of residual gas pressure

In the case of cryogenic Penning traps such as ARTEMIS, it is not possible to measure the pressure inside the trap directly. Thus, charge exchange due to collisions with a neutral gas species inside the trap acts as a probe for estimation of the residual gas pressure. Due to the cryogenic environment of the trap, majority of the gases freeze out before reaching 4 K. Hence, the major components of residual gas are helium and hydrogen (atomic and molecular).

For a thermalised cooled cloud of ions, the area under each peak can be related to the number of ions in that charge state. As depicted in section 5.6, the ion clouds in ARTEMIS can be 'cleaned' to obtain a single charge state in both CT and ST. Therefore, for such a pure ensemble of ions, the residual gas pressure p is defined by:

$$p = \frac{1}{\sigma t_c} \sqrt{\frac{k_B T \mu_m}{3}} \qquad \text{with} \qquad \mu_m = \frac{M m_r}{M + m_r} \tag{5.7}$$

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where m_r is the mass of the residual neutral gas and M is the mass of the ion under observation, σ is the electron capture cross section of the ion from the neutral residual gas, T is the temperature of the ion cloud, and k_B is the Boltzmann constant. The number of ions in a given charge state undergoes an exponential decay with time due to charge exchange. If n_0 is the number of particles at time t_0 and n_1 particles are observed at a later time t_1 , such that the decay constant (lifetime) is given as:

$$t_c = \frac{t_1}{\log(n_0/n_1)}$$
(5.8)

Figure 5.14: (a) Mass spectra of the cleaned Ar^{13+} ion ensemble trapped in ST. At t = 0, spectrum shows a cooled cloud of Ar^{13+} ions. Due to interaction with the residual gas, charge exchange takes place and at t = 18 h, lower charge states can be seen in the spectrum. (b) Exponential decay for Ar^{13+} ion number in ST with a decay constant of 5 h. The ion number is estimated from area under the peak.

For the first time, pressure inside the trap could be measured by storing the ions in the spectroscopy trap. In the figure 5.14a, the axial ion signal of Ar^{13+} ions from the q/m spectrum in ST at time t = 0 and t = 18 h have been shown. The number of ions in 13+ charge state decreases with time, whereas the ion number for the lower charge state (12+) increases, indicating charge exchange. A cooled cloud of Ar^{13+} ions is confined in ST and different spectra are recorded after a time of 2 h, 8 h and 18 h after the first scan. Calculation of area under the peak for Ar^{13+} gives an estimate on the ion number which follows an exponential decay as shown in figure 5.14b. From the graph 5.14b, the lifetime of 5 h is obtained for Ar^{13+} ions. From the Müller-Salzborn fit [41], the charge exchange cross-section is 2.7×10^{-18} m². This estimates an upper limit on the pressure to be 3.1×10^{-14} mbar for the ensemble at 4 K. It should be noted that the estimated pressure is not absolute but an upper bound on the actual pressure inside the trap. This is because the ion number is estimated by calculating the area under the peak. In a q/m spectrum, the area under the peak is a convolution of energy of the ions and number of ions. Since each voltage scan also cools the ions, hence the exponential curve decays stronger than expected only through a change in ion number. Thus, the actual pressure in the trap is better than the value estimated in this work.

Figure 5.15 shows a similar measurement for residual gas pressure obtained in CT for Ar^{11+} ions. A pure cooled cloud of Ar^{11+} ions is stored in CT and over a span of 32 hrs, various q/m spectra are acquired at intervals of 6.5 hours. The charge exchange is observed and lower charge states can be seen in figure 5.15a after t = 32.8 h. With the lifetime value of 25.7 h, a similar pressure estimate of 1.2×10^{-14} mbar is obtained as for the measurements in ST.



Figure 5.15: (a) Mass spectra of cleaned Ar^{13+} trapped in CT. At t = 0, spectrum shows a cooled cloud of Ar^{11+} ions. Due to interaction with the residual gas, charge exchange takes place and at t = 32.8 h, lower charge states of Ar^{10+} , Ar^{9+} and Ar^{8+} can be seen in the spectrum. (b) Exponential decay for Ar^{11+} ion number in CT with a decay constant of 25.7 h. The ion number is estimated from area under the peak.

Chapter 6 The SHIPTRAP experimental set-up

The Separator for Heavy Ion reaction Products (SHIP) located at the end of the UNILAC (UNIversal Linear ACcelerator) in GSI Helmholtzzentrum für Schwerionenforschung GmbH, Germany (figure 4.1), is an electromagnetic recoil separator to investigate super heavy elements. These elements are produced at SHIP, for example, using the 'cold-fusion process' [118]. SHIPTRAP is a double Penning trap experiment dedicated to high-precision mass measurements and is located behind the velocity filter at SHIP. Direct high-precision mass spectrometry calls for proper ion preparation, thermalisation, and stopping. For this reason, the fusion evaporation reaction [119] products with energies of tens of MeV from velocity filter are injected into the cryogenic gas stopping cell (CGC) of SHIPTRAP. The ions are slowed down and thermalised in the CGC (helium pressure of $7.5 \,\mathrm{mbar}$ at a temperature of $40 \,\mathrm{K}$) and further extracted through the extraction radio-frequency quadrupole (RFQ) having a helium pressure of 10^{-2} mbar. The continuous ion beam is converted to bunches using the RFQ buncher (helium pressure of 10^{-3} mbar). Finally, these cooled ion bunches are transferred to the double Penning trap set-up located inside the bore of a 7T superconducting magnet and are projected onto a detector for high-precision mass measurements. The schematic view of the SHIPTRAP set-up has been shown in figure 6.1.

6.1 The velocity filter at SHIP

Super heavy nuclides (Z > 103) are produced at SHIP through fusion-evaporation reactions. For the production using the 'cold-fusion process', the beam interacts with a rotating target wheel consisting of 8 foils of Pb or Bi. The UNILAC accelerator



Figure 6.1: Schematic view and vertically mirrored photograph of the current SHIPTRAP set-up. The cooled ions from CGC are extracted through the extraction RFQ. The continuous ion beam is thus bunched using a buncher RFQ. D1, D2, D3, and D4 show various detector positions. D4 represents the delay line position-sensitive MCP detector. D1, D2 and D3 are the detector positions with movable mounts for α -detectors and ion detectors. The figure on the top shows the schematics of the major components of the SHIPTRAP set-up and is modified from [120]. Below it is a vertically mirrored photograph of SHIPTRAP (Picture credits: G.Otto/GSI).

of GSI delivers a pulsed beam (5 ms pulses, 50 Hz) of heavy ions with energies \approx 5 MeV u⁻¹, which is sufficient to overcome the Coulomb barrier between the target nuclei and the projectile, thus forming an excited compound nucleus. This excited compound nucleus cools down by evaporation of neutrons, and the reaction product leaves the target in the direction of the beam. The target wheel is rotated in order to increase the interaction surface with the projectile beam and also to allow for cooling of the target. Due to conservation of momentum the reaction products, being heavier than the unreacted projectiles, move with a smaller velocity as compared to the primary beam. The velocity filter at SHIP exploits this fact to separate the fusion-evaporation residues (yellow line in figure 6.2) from the remaining primary beam (red line), using a special combination of electrostatic and magnetic fields. Electric deflectors and dipole magnets constitute the main part of the velocity filter.

ions with a certain velocity, given by v = E/B, to pass through the filter independent of their charge. Due to scattering in the target material, the residual beam has an angular divergence which is focused by the three electromagnetic quadrupole lenses at the beginning and the end of the separator. The last bending magnet deflects the beam by 7.5° to further reduce the background [121, 122].



Figure 6.2: Schematic view of the velocity filter at Separator for Heavy Ion reaction Products (SHIP). The filtered products (yellow line) are then transmitted towards SHIPTRAP. Figure has been taken from [122].

6.2 Cryogenic gas stopping cell and radio-frequency quadrupole buncher

The fusion evaporation products from the velocity filter at SHIP are inserted into the cryogenic gas stopping cell through optional degrader foils (thickness ranging from $0.5 \,\mu\text{m}$ to $4.5 \,\mu\text{m}$) and a titanium entrance window (thickness of a few μm depending on energy of the incoming particle beam). For high-precision Penning trap mass measurements of nuclides with lowest yield, it is important to have a high ion stopping efficiency and loss free thermalisation. This can be achieved in a cryogenic gas cell (CGC). In the SHIPTRAP CGC, the standard operating parameters are 40 K and 7 mbar of helium which corresponds to a room-temperature equivalent pressure of about 50 mbar of helium (see figure 6.1). The general requirement in a gas cell is to have minimum impurities thereby also reducing the ion-ion interaction. This

leads to a decline in the unwanted molecular ion formation, charge exchange interaction and recombination reactions. In order to have high level of purity, ultra-high vacuum conditions are achieved before filling the gas cell with highly-purified helium gas. By operating the gas cell at cryogenic temperatures, the remaining impurities are frozen out on the surfaces. The CGC has two vacuum chambers, an inner chamber and an outer chamber. The inner chamber has active buffer gas volume at a room-temperature equivalent pressure of 50 mbar. The vacuum in the outer chamber thermally isolates the inner chamber. The inner chamber has a diameter of 400 mm, a length of 450 mm and is provided with good thermal insulation using multi-layered insulation foils to reduce the heat load.



Figure 6.3: Schematic view of the inner chamber of the cryogenic gas stopping cell at SHIPTRAP.

The stopped ions from the CGC are guided towards the *de Laval* type extraction nozzle with the help of an electric field. The field is created by the DC voltages applied to 8 cylindrical electrodes (labeled as 'dc cage', figure 6.3) and the oscillating field is created by applying voltages to 76 ring type electrodes with successively reducing diameter (labeled as 'rf funnel' in figure 6.3) [123, 124]. The low pressure area of the extraction RFQ (pressure ~ 10^{-2} mbar) is connected to the high pressure area in the cryogenic gas stopping cell (room-temperature pressure equivalent ~ 50mbar) through the *de Laval type* nozzle, creating a supersonic gas jet for ion injection into the extraction RFQ. This guides the ions into the RFQ buncher and cooler (having a helium pressure of approx. 10^{-3} mbar). The continuous ion beam needs to be converted into bunches in order to efficiently trap the ions in the double Penning trap set-up. The extraction RFQ and buncher consist of axially segmented rods. A DC voltage gradient is applied in the extraction RFQ along its symmetry axis in order to guide the ions towards the buncher. The segmented rods in the buncher RFQ are subjected to DC voltages such that a potential minimum is created at the second to last segment. Detailed description of the CGC and the extraction RFQ and buncher can be found in [120, 123, 125]. For this work, two recoil-ion sources were mounted inside the cryogenic gas cell, one is the ²²⁵Ac source facing the entrance window and the other is the ²²³Ra source facing downstream towards the beamline. These sources have been described in detail in section 7.1.

6.3 Reference ion sources

For high-precision Penning trap mass spectrometry, a reference ion source providing ions with charge-to-mass ratio similar to the ion of interest is required. The SHIP-TRAP set-up has two different types of reference ion sources, a laser-ablation source capable of providing different ions with various charge-to-mass ratios and two surface ion sources. Ions are transported from the reference ion sources to the traps by a series of electrostatic ion-optics elements.

Laser-ablation ion source

For different offline measurements with various stable and long-lived reference ions, the laser-ablation ion source may be used [126, 127]. The target disk consists of multiple elements in the form of thin metallic foils or a liquid solution deposited on thin titanium foil and evaporated to dryness. It is mounted on a rotatable motor and irradiated with a pulsed 532 nm frequency-doubled Nd:YAG laser to produce reference ions. The degree of rotation of the stepper motor can be defined and fine-tuned in order to access multiple elements at a given point of time. The created ions are then transferred to the mini RFQ to axially confine the ions. The mini RFQ consists of an injection electrode, four RFQ rods, and an ejection electrode. The ions confined in the RFQ are thermalised using the buffer gas cooling technique with helium gas (section 2.5.1). The cooled reference ions are then transferred to the Penning trap by lowering the potential at the ejection electrode.

Surface ion sources

The stable reference ion sources with Cs and Rb ions are mounted off-axis (figure 6.1) relative to the beam direction and the ions are efficiently transported to the trap using the electrostatic ion-optics elements. These are surface ionisation sources. In

such a source, when a sufficiently high current is passed through a filament, the metal surface is heated up. The resulting temperature of the backing metal is high enough to cause desorbtion of ions (or atoms) of the metal of interest. The ionisation happens when the ionisation potential of the targeted atoms (e.g Cs and Rb in this case) is less than the work function of the backing metal. Since alkali and alkaline earth metals have low ionisation potentials (of the order of $\sim 5 \text{ eV}$), they are ideal candidates for the surface ion source. The current passing through the filament regulates the temperature of the filament and in turn the number of ions produced, whereas the energy of the ions is dependent on the voltage applied to the filament. Ion pulses are created by switching a deflector.

6.4 Dual Penning trap set-up

SHIPTRAP uses a dual Penning trap set-up located inside the bore of a 7 T superconducting solenoid magnet. The set-up consists of two traps, Preparation Trap (PT) and Measurement Trap (MT) as shown in figure 6.4. The preparation trap is used for ion preparation and isobaric purification of the ion samples, featuring a typical resolving power of 10^5 or better. The second trap is the measurement trap which allows for ion manipulation by exciting the eigenmotions of the ion of interest. Both PT and MT at SHIPTRAP have electrically compensated open-endcap cylindrical Penning trap geometries (see section 2.1).

The cylindrical electrodes forming the SHIPTRAP Penning traps are made up of gold-plated oxygen-free high-conductivity copper (OFHC) and the electrical insulation between the conducting electrodes is provided by aluminium oxide (alumina). These materials have a negligible influence on the homogeneity of the high magnetic field. The electrodes are each 8 mm in length and are separated by 0.5 mm. The Preparation trap (PT), into which the ions are injected, is 212 mm long while the second trap, i.e. the Measurement Trap (MT), is 185 mm in length. Both of the traps have an inner diameter of 32 mm. The ring electrodes for each of these traps are positioned such that they lie at the two homogeneous magnetic field regions in the magnet and are separated by 200 mm. Both of the ring electrodes are azimuthally segmented into eight parts in order to be able to provide rf excitation in different geometries to the ions [128]. On either side of the ring electrodes in PT and MT, there are correction electrodes and a three-fold axially segmented endcap electrode. The endcap electrode is segmented axially in order to be able to apply different DC



Figure 6.4: Schematic view of the dual Penning trap set-up at SHIPTRAP. The traps are located inside a 7 T magnet bore. Figure modified from [56].

potentials at each segment¹. This would help to create an extended potential well for an even more efficient capture of ions. The PT has one pair of two-fold segmented electrodes and one pair of non-segmented electrode on each side of the central ring electrode, whereas the MT has a pair of two-fold azimuthally segmented correction electrodes. The correction electrodes are segmented so that a quadrupolar excitation can be applied to the ions which couples the axial oscillation with one of the radial modes of the ions' motion.

The traps are separated by a 52 mm long diaphragm having a diameter of 1.5 mm which acts as a pumping barrier. In order to provide access to helium gas in the PT for buffer gas cooling, one of the endcaps has an inlet which helps maintain a buffer gas pressure of $\sim 5 \times 10^{-5}$ mbar. For more details regarding the geometry and construction of Penning traps at SHIPTRAP refer to [129, 130, 131].

6.5 Detection system

In order to monitor and optimise ion transmission efficiency, various detectors have been mounted at multiple locations throughout the SHIPTRAP beamline. As indicated in figure 6.1, detectors are mounted at positions D1, D2, D3, and D4. At the detector positions D2 and D3, ion detectors, α detectors and ion optics elements are

¹This feature of the endcap electrode is currently not utilised.

mounted on movable feedthroughs such that they can be used interchangeably. The ion detectors (channeltron or micro channel plate (MCP) detector) are installed to count the number of ions or to measure the time-of-flight distribution of the extracted ions. In order to obtain the position and time-of-flight of the ions extracted from MT, a position-sensitive delay line detector is placed at D4.

The α detectors

The α detectors are installed in the SHIPTRAP set-up to measure the extraction efficiency and to record α -spectra from radioactive ions. The α particles emitted by the radioactive ions in the experiment create electron-hole pairs in the silicon α detectors, which in turn creates an electric current proportional to the energy of the particle. The ions extracted from the CGC cannot be implanted directly onto the surface of silicon α detector due to their low energy and hence, a 0.5 µm thick aluminium foil, biased at ~1.5 kV, is placed in front of the detector (figure 6.5). The energy resolution for the α detector ranges from few tens of keV to about 100 keV. Radioactive isotopes can be identified by their characteristic decay energy.



Figure 6.5: The time-of-flight MCP detector, α detector and ion optics mounted on a movable feedthrough. The image on the bottom right side depicts the position-sensitive delay line detector (DLD40 by ROENTDEK) installed after the traps [56].

Ion detectors

In the SHIPTRAP set-up, channeltron and MCP detectors are installed in order to record the time-of-flight spectra and to measure the number of ions. A channeltron detector (SJUTS KBL25RS) and a chevron MCP detector (TOPAG MCP-MA25/2) are placed at position D2 and D3, respectively. The time-of-flight information obtained from these detectors can provide an information on the charge-state distribution of ions extracted from the RFQ buncher (or the PT/MT). For fast repetitive measurements and optimisation, channeltron detectors are used at the cost of ion selectivity. The working principle of both these detectors is based on electron multiplication by secondary electron emission. An ion entering one of the channels in an MCP hits the surface of the detector and produces secondary electrons. Due to a potential gradient across the channels, these secondary electrons further hit the internal walls of the channel and lead to a cascade of electrons. These electrons are collected at the anode, giving a large detectable signal. The chevron MCP stack is an assembly of two MCP plates, stacked together with their micro channels at a certain angle. In this way, large number of secondary electrons are produced because the electrons exiting the first plate, cascade in the second plate. By introducing an angle between the channels, higher gain is produced at a given voltage and the ion feedback is reduced. The MCP detector has multiple channels while the channeltron detector has only one channel (also known as CEM: Channel Electron Multiplier).

Delay line detectors

For high-precision mass spectrometry using the PI-ICR technique, a position-sensitive delay line detector is installed behind the traps at a distance of 80 cm from the center of the MT. A delay line detector, DLD40 from ROENTDEK, consists of two MCPs in chevron configuration and a delay line anode. The delay line anode consists of two layers of wires oriented perpendicular to each other. This enables determination of position in two dimensions, one each for measurement in x- and y-direction. For each layer, the position information is encoded by the time difference in the signal arrival on both ends of delay lines while the MCP signal provides time-of-flight information. The detector has an active diameter of 42 mm, a position resolution of 70 μ m and a time resolution of 10 ns. The efficiency of the detector is about 30 %.

Chapter 7 Mass measurements at SHIPTRAP

The SHIPTRAP Penning trap experiment is dedicated to the direct high-precision mass measurements. In this work, measurements were performed on nuclides from two radioactive ion sources installed in the cryogenic gas cell. These measurements were carried out using the phase-imaging ion-cyclotron-resonance technique (PI-ICR, section 3.6). The raw data obtained during the measurements was analysed by determining the cyclotron frequency of the ion of interest and the reference ion. This was followed by the calculation of the mean frequency ratios. A number of systematic uncertainty sources were investigated. The final results obtained for the first-ever direct mass measurements for some of the decay products of the recoil-ion sources are compared to the literature values provided in the Atomic Mass Evaluation 2020 [88].

7.1 Recoil-ion sources 223 Ra and 225 Ac

The recoil-ion sources were installed in the cryogenic gas cell (CGC, figure 6.1). These α emitters act as offline sources to characterise and optimise the CGC. In an α -decay, the parent nucleus decays into a daughter nucleus and an alpha particle, while conserving energy and momentum in the process. For α -decay in nuclei with $A \approx 200$ and $Q_{\alpha} \approx 6$ MeV, the daughter nucleus has high kinetic energy (~ 75 keV). This energy is enough for the daughter nucleus to leave the source holder material. This also imparts the ion source its name 'recoil-ion source'. In the cryogenic gas cell, the ions emitted from these point-like recoil sources are stopped and thermalised due to the presence of helium buffer gas. The characteristic α -decay energy of the emitted daughter nuclei helps to identify the ions extracted from the gas cell.

Not every sample of radioactive material can be used as a recoil-ion source. In order to qualify as a recoil-ion source at SHIPTRAP, it should fulfill the following conditions:

7. Mass measurements at SHIPTRAP

- The recoil-ion source is installed in the cryogenic gas cell at SHIPTRAP, which has stringent cleanliness requirements (section 6.2). Thus, the ion source should not contain any impurities such as organic solvents during ion source fabrication or long-lived radio-nuclides, to avoid contamination of the CGC.
- It should have a sufficiently high activity for the parent nucleus. Although the SHIPTRAP experiment is sensitive enough for count rates of a single ion per day, higher count rates are favorable to characterise and optimise the CGC.
- The process of ion source production, transportation and finally installation in the CGC takes a couple of days. For the purpose of calibration, recoil-ion sources are also utilised in the CGC during online experiments. This further necessitates a parent nucleus with an even longer half life (based on the activity and required count rate) since the online experiments can last several weeks.
- The half-life of the first recoiling daughter must be longer than the CGC's extraction time of tens of milliseconds, but short enough to allow for fast repetitive measurements, specifically for measurements of the extraction efficiency of the gas cell. Additionally, the decay chain must not contain any radioactive isotope with a very long half-life to avoid the contamination of the gas cell and deposition of long-lived radioactive isotopes on the position sensitive detector.
- To ensure selective and sensitive detection, the recoiling daughter ion should also be an α emitter. Moreover, none of the α energies of the decay chain should overlap with the α energies of the particle of interest during online experiments.

During the present experimental campaign for high-precision mass measurement, the off-line recoil-ion sources ²²³Ra and ²²⁵Ac were mounted inside the CGC. The decay schemes of these are shown in figure 7.1. These ion sources have been produced at the Institute for Nuclear Chemistry Mainz by collecting in the gas phase. Ion source production by molecular plating leads to the formation of a dead layer. The dead layer formation is due to the deposition of organic impurities on the source holder [133, 134, 135, 136]. On the contrary, the recoil-ion sources produced in gas phase do not have a significant dead layer. This is due to the clean and controllable conditions under which they are produced.

The alpha spectra shown in figure 7.2 have been measured using a MIRION-PIPS-A450-18AM α -detector placed in vacuum at a distance of $\approx 23 \text{ cm}$ from the source. The detector has an active area of 450 mm^2 [137]. The activity of the parent



Figure 7.1: Decay scheme of the recoil-ion sources (a) 223 Ra and (b) 225 Ac [132].

nucleus changes during the measurement, but this can be neglected because the time of measurement is much smaller than the half-life of the parent. The calculated initial activities of the recoil-ion sources ²²³Ra and ²²⁵Ac are shown in table 7.1. The activity of the parent nucleus at any given time t_0 , placed at a distance R_d from an α -detector with effective surface area, $\pi (d_{det}/2)^2$ is given by:

$$A(t_0) = \frac{A_{meas}}{\Omega\mu_{branch}}, \quad \text{with} \quad A_{meas} = \frac{\alpha_{parent}}{t_{meas}} \quad \text{and} \quad \Omega = \frac{\pi (d_{det}/2)^2}{4\pi (R_d)^2}$$
(7.1)

where $A(t_0)$ is the absolute initial activity of the source, μ_{branch} is the α branching ratio, A_{meas} is the measured activity of the parent and t_{meas} is the measurement time. A_{meas} can be obtained from figure 7.2 by calculating the area under the peak, which gives the number of decays. Ω is a geometric factor representing the solid angle between source and α -detector with an active surface diameter of d_{det} .

Table 7.1: Initial radioactivity of recoil-ion sources 223 Ra and 225 Ac used in the present work at SHIPTRAP.

Parent nucleus	Initial radioactivity	Measurement duration
²²³ Ra	$3\mathrm{kBq}$	1 h
²²⁵ Ac	$150\mathrm{Bq}$	13 h

As can be seen from the decay chains of ²²³Ra and ²²⁵Ac in figure 7.1, the halflives of ²¹⁵Po, ²¹⁷At and ²¹³Po are of the order of few milliseconds or lower. Therefore,



Figure 7.2: Calibration spectra from recoil-ion sources ²²³Ra and ²²⁵Ac.

masses for these radio-nuclides could not be measured. Furthermore, due to the low yield of ²¹¹Bi, ²¹¹Po and ²⁰⁹Tl, mass measurements could not by performed for these nuclides. Within the scope of this work, masses of ²²¹Fr, ²¹⁹Rn, ²¹³Bi, ²¹¹Pb, ²⁰⁹Pb, ²⁰⁷Pb and ²⁰⁷Tl are measured.

7.2 Data analysis

This section briefly describes the analysis process for the data obtained from the recoilion sources. The measurements were mostly made by using the double-pattern PI-ICR technique (described in section 3.6.2). The analysis begins with the suppression of the background in the raw data, then the cyclotron frequencies of the ion of interest and the reference ion are determined. This is followed by a determination of frequency ratios. Individual frequency ratios are calculated using interpolation of the reference ion frequency at the time of the ion of interest's measurement. These frequency ratios are later averaged, using the individual uncertainties as weights. A number of systematic uncertainty sources are investigated and included in the results along with the statistical uncertainty.

At several points it will be required to estimate the value of a quantity X at a certain magnitude (t_i) when the measurement is done at a different magnitude (e.g. the reference ion's cyclotron frequency at the time of the ion of interest's frequency measurement). To do this, linear interpolation is used. The interpolated quantity X is then:

$$X^{lin}(t_i) = X(t_{i-1}) + \left(X(t_{i+1}) - X(t_{i-1})\right) \left(\frac{t_i - t_{i-1}}{t_{i+1} - t_{i-1}}\right)$$
(7.2)

and the associated uncertainty is given by:

$$\delta X^{lin}(t_i) = \sqrt{\left(\frac{t_i - t_{i-1}}{t_{i+1} - t_{i-1}}\delta X(t_{i+1})\right)^2 + \left(\left(1 - \frac{t_i - t_{i-1}}{t_{i+1} - t_{i-1}}\right)\delta X(t_{i-1})\right)^2}$$
(7.3)

In some cases, the error is obtained by comparing the deviation of the known values to the interpolated values as in the case of non-linear magnetic field drift effects in section 7.2.4.

7.2.1 Raw data

The starting point of the data analysis for measurements with the ²²³Ra and ²²⁵Ac radioactive sources is in the form of two distinctly measured phase images for the PI-ICR technique. These images show the final phases in the magnetron and modified cyclotron modes (defined in equation 3.16). In order to distinguish between the background and the actual signal, time-of-flight and position gates are applied. The final spots after applying these gates have been depicted in figure 7.3. The background arises due to dark counts of the detector or due to the presence of unwanted ion species. The ion occurrences lying within two standard deviations of the mean value are further analysed.

7.2.2 Determination of the cyclotron frequency

As explained in section 2.2, for high-precision mass measurements using the PI-ICR technique (section 3.6.2) the cyclotron frequency of an ion can be determined by:

$$\nu_c = \frac{\omega_c}{2\pi} = \frac{\phi + 2\pi(n_+ + n_-)}{2\pi t_{acc}}$$
(7.4)

where $\phi = \phi_{-}^{final} - \phi_{+}^{final}$ is the angle difference between the magnetron and modified cyclotron phase accumulated during the time t_{acc} . n_{+} and n_{-} are the number of full



Figure 7.3: Final phase spots of the magnetron and modified cyclotron modes after applying the time-of-flight gate and position gate for the 207 Pb / 207 Tl mass doublet. The images are obtained from the position-sensitive delay-line detector at the D4 position (figure 6.1). The position of the trap center is represented by a cross (x). Both of the spots are obtained by implementing the PI-ICR technique described in section 3.6.

revolutions completed during t_{acc} . Being an integer number, they do not contribute to the statistical uncertainty in the frequency measurements. The accumulation time is determined by the frequency generator which is locked to a 10 MHz rubidium clock. Due to the low relative uncertainty (better than 10^{-11} over 1 s) of the rubidium frequency standard, the error in t_{acc} is neglected. The uncertainty in the measurement of ν_c is thus only dependent on the uncertainty in the phase and is given by:

$$\delta\nu_c = \frac{\delta\phi}{2\pi t_{acc}}\tag{7.5}$$

As seen in equation 7.4, in order to obtain the cyclotron frequency of the ion, we need to calculate the phase difference accumulated in the time t_{acc} . A determination of the coordinates of the center, magnetron and modified cyclotron projected spots, gives the angle for each of the spots. As shown in figure 7.4, the phase spots and center are projected on the x-y axes, and Gaussian fits give the x-y coordinates and the corresponding fit uncertainties (δx_i and δy_i), such that:

absolute position coordinates = $(x_i \pm \delta x_i, y_i \pm \delta y_i) \quad \forall \quad i \in [\text{center}, -, +]$ (7.6)



Figure 7.4: (a) Projection of the phase spot on x- and y-axis and phase angle determination of the spots with reference to the center spot. As the accumulation time t_{acc} increases, ϕ_{-} decreases and ϕ_{+} increases, as depicted by the dotted lines in the figure. (b) Gaussian fits to the projected coordinates give the absolute position of the spots shown in (a).

Due to fluctuations of the electric field, the positions of the center spots can change over time which leads to a systematic uncertainty (section 7.2.4). Thus, in order to define the trap center at the measurement time of the ion of interest's frequency (t_i) , two center measurements are taken at t_{i-1} and t_{i+1} such that $t_{i-1} < t_i < t_{i+1}$.¹ The coordinates of the two center measurements are thus linearly interpolated at the time t_i , as per equation 7.2.

For $\bar{\xi}_{\pm} \equiv (\xi_{\pm} - \xi_{center})$ and uncertainties $\delta \bar{\xi}_{\pm} = \sqrt{(\delta \xi_{\pm})^2 + (\delta \xi_{center})^2}$, where $\xi \in [x, y]$, the relative positions are defined as:

relative magnetron spot =
$$(\bar{x}_{-} \pm \delta \bar{x}_{-}, \bar{y}_{-} \pm \delta \bar{y}_{-}),$$

relative modified cyclotron spot = $(\bar{x}_{+} \pm \delta \bar{x}_{+}, \bar{y}_{+} \pm \delta \bar{y}_{+}).$ (7.7)

The phase angles of the magnetron spot and the cyclotron spot are given by:

$$\phi_{\pm} = \begin{cases} \operatorname{atan2}(\bar{y}_{\pm}, \bar{x}_{\pm}), & \text{for } \operatorname{atan2}(\bar{y}_{\pm}, \bar{x}_{\pm}) \ge 0\\ 2\pi - |\operatorname{atan2}(\bar{y}_{\pm}, \bar{x}_{\pm})|, & \text{for } \operatorname{atan2}(\bar{y}_{\pm}, \bar{x}_{\pm}) < 0 \end{cases}$$
(7.8)

The uncertainty in the phase angles is calculated using the Gaussian propagation of uncertainty (GPOU) :

$$\delta\phi_{\pm} = \sqrt{\frac{(\bar{x}_{\pm}\delta\bar{y}_{\pm})^2 + (\bar{y}_{\pm}\delta\bar{x}_{\pm})^2}{\bar{x}_{\pm}^2 + \bar{y}_{\pm}^2}} \tag{7.9}$$

¹The center spots are recorded in a separate measurement file than the phase spot measurement. At a given point of time either a double pattern or a center spot measurement is performed. Therefore, the center cannot be measured at t_i and the coordinates need to be interpolated.

7. Mass measurements at SHIPTRAP

The phase difference ϕ between the absolute phase angles of the magnetron and modified cyclotron spot is given by:

$$\phi = \phi_{-} - \phi_{+} \tag{7.10}$$

and the uncertainty is given by:

$$\delta\phi = \sqrt{(\delta\phi_-)^2 + (\delta\phi_+)^2} \tag{7.11}$$

With this, the measured true cyclotron frequency ν_c for the reference ion and the ion of interest can be obtained by making use of equations 7.4 and 7.5.

7.2.3 Mean cyclotron frequency ratio determination

In order to measure the mass of an ion, phase spot measurements are done for a reference ion along with the ion of interest. This eliminates the dependence of the ion's mass on the magnetic field and facilitates higher precision. The mass of the desired ion is proportional to the cyclotron frequency ratio $R = \nu_c^{ref}/\nu_c^{ion}$. The frequencies of the reference ion and the ion of interest are measured alternatively during the measurements at SHIPTRAP. This alternation is necessitated by the temporal variation of the magnetic field. If the field was constant, there would not be any need to alternate and a single, very long measurements could be made for the reference ion and the ion of interest. However, due to the changes in the magnetic field, the measurements are spaced out over time. Therefore, the frequency of reference ion is interpolated at the time of measurement of ion of interest according to equations 7.2 and 7.3 with $X = \nu_c^{ref}$, giving $\nu_c^{ref,lin}(t_i)$ and the associated uncertainty $\delta \nu_c^{ref,lin}(t_i)$.

The cyclotron frequency ratio at time t_i and uncertainty associated with this ratio are, respectively, given by $R(t_i)$ and $\delta R(t_i)$:

$$R(t_i) = \frac{\nu_c^{ref,lin}(t_i)}{\nu_c^{ion}(t_i)},\tag{7.12}$$

$$\delta R(t_i) = \sqrt{\left(\frac{\delta \nu_c^{ref,lin}(t_i)}{\nu_c^{ion}(t_i)}\right)^2 + \left(\frac{\nu_c^{ref,lin}(t_i)\delta \nu_c^{ion}(t_i)}{(\nu_c^{ion}(t_i))^2}\right)^2 + \xi_B^2 R^2(t_i) + \xi_D^2 R^2(t_i)} \quad (7.13)$$

where ξ_B is the systematic uncertainty due to the non-linear magnetic field drift and ξ_D is the uncertainty due to distortion of phase images. The contribution due to ξ_D is typically of the order of 10^{-10} [138], which is lower than the current statistical uncertainty of > 10^{-9} , and hence can be neglected.

The mean cyclotron frequency ratio (R_{mean}) of the measurement set is calculated from the error-weighted arithmetic mean of N single frequency ratio measurements, such that:

$$R_{mean} = \frac{\sum_{N} w_i R(t_i)}{\sum_{N} w_i} \text{ with } w_i = \frac{1}{(\delta R(t_i))^2}$$
(7.14)

The statistical error in the measurement data set is categorised as internal and external error. The standard error of the weighted mean value is referred as the internal error, and the biased weighted mean variance represents the external error. These are, respectively, given by:

$$\delta R_{int} = \sqrt{\frac{1}{\sum_{N} w_i}}, \qquad \delta R_{ext} = \sqrt{\frac{\sum_{N} w_i (R(t_i) - R_{mean})^2}{(N-1)\sum_{N} w_i}}$$
(7.15)

Tha majority of systematic uncertainties in Penning trap mass spectromentry are well known (described in section 7.2.4). In order to accommodate any inconsistencies due to unknown effects in the measured data set, the internal error is adjusted using the 'Birge method' [139]. When the data set is inconsistent, the estimate for the internal error given in equation 7.15 should not be used. Rather, the associated uncertainties to each of the values should be multiplied by a common factor, which is the 'Birge ratio' (σ_B);

$$\sigma_B = \sqrt{\frac{1}{N-1} \sum_{N} \frac{(R(t_i) - R_{mean})^2}{(\delta R(t_i))^2}}$$
(7.16)

such that:

$$\delta R'(t_i) = \sigma_B \times \delta R(t_i) \tag{7.17}$$

and

$$\delta R'_{int} = \sigma_B \times \delta R_{int} \tag{7.18}$$

where $\delta R'(t_i)$ is the adjusted error in each measurement and $\delta R'_{int}$ is the adjusted internal error. Generally, when the ratio of external and internal errors is less than 1, the scattering of data is statistical and the uncertainties are jointly overrated. In the case when the ratio is more than 1, it indicates the presence of additional systematic uncertainties.

The final uncertainty in the mean cyclotron frequency ratio is given by:

$$\delta R_{mean,total} = \sqrt{\delta R_{int}^{\prime 2} + \delta R_{1,mean}^2 + (\delta_{syst} R_{mean})^2}$$
(7.19)

where $\delta R_{1,mean}$ is the mass dependent systematic uncertainty and δ_{syst} is a factor which accounts for additional unknown systematic uncertainties [140]. These systematic uncertainties have been discussed in detail in section 7.2.4.

7.2.4 Systematic uncertainties

Center spot scattering

The center of the measurement trap is projected on the position sensitive detector as the center image spot. The center spot for the magnetron and modified cyclotron mode is obtained without applying any excitation (as in figure 3.9). The x-y coordinates of the center spot are obtained by a Gaussian fit. The position of the center spot can change over time [89], and thus must be interpolated at the time of the phase spot measurement. The absolute center position at the phase spot measurement time t_i are given by linear interpolation of the consecutive center measurements made at t_{i-1} and t_{i+1} using equation 7.2 with $X \in [x,y]$.

The coordinates of the center spot are observed to scatter around the mean value when plotted with respect to relative time² (figure 7.5). To include the effect of scattering of the center spot in the data analysis, the standard deviation of the center spots' coordinates is taken as their uncertainty. For N center measurements with $\xi_{mean,center}$ as the mean center, this uncertainty is given by:

$$\delta\xi_{center} = \sqrt{\frac{1}{N-1} \sum_{N} (\xi_{center}(t_i) - \xi_{mean,center})^2} \quad ,\xi \in [x,y]$$
(7.20)

The standard deviation of the centers is calculated individually for each ion species by using their respective centers.

Mass dependence of the center position

When the coordinates of the individual center measurements are plotted with respect to the overall mean center (figure 7.5) it becomes clear that the center image for different species is at a slightly different position. This is more pronounced in the y-axis. In order to get an estimate of the mass-dependence of the centers on the final result, the analysis was performed in two different ways:

- a) by interpolating the center of the reference ions at the time of the phase spot measurement of ion of interest
- b) by interpolating the center of the respective ions at the time of the phase spot measurement.

 $^{^{2}}$ A scattering in the centers' coordinates is expected within their error bars. However in the present case, they scatter beyond what their error bars would suggest.



Figure 7.5: Scattering of centers of different ion species around the mean value. The time of each value is taken relative to the beginning of the measurement campaign. The center spots are obtained by using the PI-ICR technique with $t_{acc} = 1200 \text{ ms}$ and the excitation amplitude set to 0 V. In figure (b), a stronger mass dependent shift can be observed in the y-coordinate of the center spot. In the inset, the scattering of the spots with mass values 197 and 219 is shown.

The final result is found not to be significantly different in both these cases. Therefore, the reference centers are used as target ion centers in the cases where no individual center measurements were performed.

In figure 7.5, a mass dependent shift in the center position has also been observed. The origin of this mass-dependence of the center position remains unexplained so far.

Non-linear magnetic field drift

The linear interpolation of the frequency of the reference ion is only a first-order estimate of how the magnetic field changes over time. Thus, non-linear magnetic field fluctuations have to be taken into account. This is done by evaluating the relative standard deviation for a set of N consecutive and equally spaced frequency measurements. The non-linearity of the magnetic field drift $\Delta B = B - B^{lin}$ is calculated by taking the difference between the measured cyclotron frequency ν_c and linearly interpolated frequency value (ν_c^{lin} , equation 7.2).



Figure 7.6: Plot of the relative standard deviation of magnetic field with respect to the interpolated value as a function of time intervals ΔT between the two ¹³³Cs⁺ measurements. The slope accounts for time-dependent non-linear magnetic field systematic uncertainty. Figure adapted from [89].

This process has been performed in [57, 141] and the current data corresponds to the work from [56, 89]. For a time ΔT between two successive measurements, the standard deviation is:

$$\xi_B = \sigma \left(\frac{B - B^{lin}}{B}\right) (\Delta T) = \sigma \left(\frac{\nu_c - \nu_c^{lin}}{\nu_c}\right) (\Delta T) = \sqrt{\frac{\sum_N (r(t_i) - r_{mean})^2}{N - 1}}$$
(7.21)

where r_{mean} is the error weighted average value (equation 7.14) with $r(t_i)$ defined as:

$$r(t_i) = \frac{\nu_c(t_i) - \nu_c^{lin}(t_i)}{\nu_c(t_i)}.$$
(7.22)

The standard deviation of the relative errors due to linear interpolation of the magnetic field, measured with respect to the interpolated value, is plotted as a function of time intervals ΔT (figure 7.6). The data is fitted to a linear function:

$$\xi_B \equiv \Delta T \times \delta_B \tag{7.23}$$

The slope of this curve gives the non-linear component of the temporal magnetic field fluctuations $\delta_B = 1.3 \times 10^{-9} \,\mathrm{h^{-1}}$. This temporal uncertainty ξ_B is added in quadrature to the statistical uncertainty of individual frequency ratios (equation 7.13).
Mass dependent systematic uncertainty

Imperfections in the electric field or a misalignment between the electric and magnetic field cause a shift in the ion's eigenfrequencies [142], as detailed in section 2.3. These shifts cancel out when calculating the true cyclotron frequency using the invariance theorem (equation 2.13). However, they do not cancel out when using the sideband method (equation 2.12). The cyclotron frequency, as determined by the sideband method (ν_c), deviates from the true cyclotron frequency $\nu_{c,true}$ by a factor $\Delta\nu_c$:

$$\nu_c = \nu_{c,true} + \Delta \nu_c \tag{7.24}$$

where the difference is given by [142]:

$$\Delta \nu_c \approx \nu_- \left(\frac{9}{4}\theta^2 - \frac{1}{2}\epsilon^2\right). \tag{7.25}$$

Here, θ is the misalignment angle between the electric and magnetic fields and ϵ is the harmonic distortion factor. Therefore the corrected frequency ratio in terms of the measured cyclotron frequencies is given by:

$$R_{corr} = \frac{\nu_c^{ref} - \Delta\nu_c}{\nu_c^{ion} - \Delta\nu_c} \tag{7.26}$$

Using Taylor expansion, R_{corr} can be written as:

$$R_{corr} = R + \frac{\nu_c^{ref} - \nu_c^{ion}}{(\nu_c^{ion})^2} \Delta \nu_c = R + R_1$$
(7.27)

It can be observed from the above equation that a difference in the mass (hence the cyclotron frequency) between the reference ion and the ion of interest leads to a non-zero correction factor R_1 , thereby incurring in a systematic uncertainty.

The weighted average for R_{corr} is defined in the same way as for R in equation 7.14. However, if we include $\Delta \nu_c$'s uncertainty into the individual errors, the contribution of $\Delta \nu_c$ to the final error will be reduced. In this work, this problem is circumvented by averaging the uncorrected ratios (R_{mean} , equation 7.14) and the correction is included at the final step using the Taylor expansion (equation 7.27). This gives the final corrected ratio as:

$$R_{corr,mean} = R_{mean} + R_{1,mean} \tag{7.28}$$

where $R_{corr,mean}$ is the average corrected ratio. The error corresponding to the average corrected ratio is thus defined as:

$$\delta R_{corr,mean} = \sqrt{(\delta R_{mean})^2 + (\delta R_{1,mean})^2}$$
(7.29)

7. Mass measurements at SHIPTRAP

where $\delta R_{mean} = \delta R'_{int}$ defined in equation 7.18 and $\delta R_{1,mean}$ is defined as:

$$\delta R_{1,mean} = \sqrt{\left(\frac{\partial R_{1,mean}}{\partial \nu_{c,mean}^{ion}} \delta \nu_{c,mean}^{ion}\right)^2 + \left(\frac{\partial R_{1,mean}}{\partial \nu_{c,mean}^{ref}} \delta \nu_{c,mean}^{ref}\right)^2 + \left(\frac{\partial R_{1,mean}}{\partial (\Delta \nu_c)} \delta (\Delta \nu_c)\right)^2} \tag{7.30}$$

The correction factor $R_{1,mean}$ includes the mass dependent systematic with the corresponding uncertainty $\delta R_{1,mean}$. From the measurements conducted on stable ions, the value of frequency shift $\Delta \nu_c$ is found to be 5.8(35) mHz [143] and is utilised for the correction of ratios for the present work.

Residual uncertainties

Apart from the sources of uncertainties mentioned above, the fluctuations in trap conditions such as temperature of the magnet bore, pressure inside the liquid He dewar, switchable power supply stability conditions and detector axis alignment, contribute to systematic uncertainty in the mass measurement. Both the magnet bore temperature and the pressure inside the helium dewar are actively stabilised in the SHIPTRAP set-up [144]. Figure 7.7 shows the variation of the temperature of the magnet bore, temperature of the electronics and the pressure inside helium dewar for the entire duration of the measurement campaign. Unfortunately, the pressure stabilisation was not working effectively during the measurements, as can be seen from the figure.

After the completion of the measurement campaign, frequency measurements were performed with ¹³³Cs to observe the variation of its cyclotron frequency as a function of the bore temperature as well as the helium pressure [143]. As a result, a change in the cyclotron frequency of $\sim 220 \text{ mHz/K}$ and 8 mHz/mbar was obtained. It can be seen in figure 7.7 that the magnet bore temperature is stabilised during the measurements. The maximum variation in the temperature is $\sim 20 \text{ mK}$, which is very small and hence its contribution to the total uncertainty can be neglected. However, the difference between the two extremes of the pressure curve is relatively large, $\sim 15 \text{ mbar}$. In addition to this, although the frequency is observed to vary linearly with pressure [143], the change in pressure itself is not linear. Since the non-linear magnetic field drift measurements were also performed with a non-stabilized pressure, the systematic due to non-linear changes in pressure is included there. Hence, the effect of these uncertainties on the final error budget can be neglected within the limits of achieved statistical uncertainty. It is expected that the unknown systematics would become manifest when calculating the Birge ratio, as defined in section 7.2.3.



Figure 7.7: Variation of the temperature of the magnet bore, temperature of the electronics and pressure inside the liquid He vessel.

7.3 Results and discussion

In this work, high-precision mass measurements are performed for various elements obtained from the recoil-ion sources ²²³Ra and ²²⁵Ac mounted at the CGC in SHIPTRAP (section 7.1). The atomic masses of these elements have been measured directly for the first time. These measurements were performed using the PI-ICR technique, described in section 3.6. The mass of the ion of interest has been measured by comparing its cyclotron frequency to that of a reference ion. The choice of the reference ion depends on *a*) closeness of its q/m value to the ion of interest and *b*) accuracy with which the mass value is known. In this work, the most used reference ion is ¹³³Cs⁺ for the ion species of interest in the 2+ charge state. In one case where the decay chain populates a mass doublet, the two ions are also measured against each other.

In the following sections, obtained frequency ratios and the measured atomic masses of ²²¹Fr, ²¹³Bi and ²⁰⁹Pb, from the decay chain of ²²⁵Ac, and ²¹⁹Rn, ²¹¹Pb, ²⁰⁷Tl and ²⁰⁷Pb, from the decay chain of ²²³Ra have been shown. The obtained mass excess of each of these elements is compared to the previous experimental data and the values given in the Atomic Mass Evaluation 2020 [88]. The previous experimental data provides the reaction energies, from which the mass excess can be derived using the procedure shown in section 3.7. For simplicity, the mass excess is represented as keV, instead of keV/c^2 , in the following text. In the following sections, regarding the mass measurements, the literature values for mass (µu), mass excess (keV) and their uncertainties have been obtained from the most recent Atomic Mass Evaluation (AME 2020 [88]). The individual frequency ratios in each case are plotted with respect to relative time, where the zero on the time axis corresponds to the beginning of the respective measurement sets. The plots also show the 'unadjusted' internal and external errors; however, for the final uncertainty estimation, the 'Birge adjustment' (defined in section 7.2.3) is implemented. The total error in the measurement, which is a combination of systematic and statistical errors, is given by the value in brackets.

7.3.1 Mass of ²²¹Fr

The first experiment adopted in Atomic Mass Evaluation concerning the measurement of nuclear structure and properties of ²²¹Fr has been reported by Walen *et al.* in 1962 [145]. The current mass value of ²²¹Fr, as given in Atomic Mass Evaluation 2020 [88], comes from the study of the decays ²²⁵Ac(α)²²¹Fr and ²²¹Fr(α)²¹⁷At, with a weight of 79.1% and 20.9%, respectively. Even though several decay spectroscopy experiments



Figure 7.8: (a) Individual frequency ratios of 221 Fr²⁺ with respect to the reference ion 133 Cs⁺ as a function of relative time. R_{mean} has a value of 0.8314710318(29), where the value in brackets is a combination of systematic and statistical errors. (b) Comparison of the individual mass excess values for 221 Fr derived from decay spectroscopy experiments [145, 146, 147, 148]. The plot also shows the mass excess obtained from AME 2020 [88]. The right-most value is obtained in this work, and the shaded region is its measurement uncertainty.

have been performed [145, 146, 147, 148], having Q_{α} uncertainty values of the order of ~2 keV, the uncertainty in the mass excess of ²²¹Fr is still about a factor of two higher than the uncertainty in the Q_{α} value. This is because the atomic mass of ²²¹Fr is affected by the large uncertainty in the atomic mass values of the daughter ²¹⁷At (or parent ²²⁵Ac), whose uncertainties also contribute to the final uncertainty as shown in section 3.7. Various mass excess values obtained from the decay spectroscopy experiments [145, 146, 147, 148] and the literature value listed in AME 2020 are shown in figure 7.8b in historical order (increasing time from left to right).

The first high-precision mass measurement of ²²¹Fr is presented in this work. The measurements were performed with the PI-ICR technique using ²²¹Fr²⁺ relative to the reference ion ¹³³Cs⁺ for two different accumulation times of ~500 ms and ~1200ms. The frequency ratios of ²²¹Fr²⁺ with respect to the reference ion ¹³³Cs⁺ obtained using PI-ICR are shown in figure 7.8a. These give a mean frequency ratio R = 0.8314710318(29) with a relative uncertainty of $\delta R/R = 3.5 \times 10^{-9}$. The final uncertainty given here is calculated using equation 7.19 with the Birge adjusted internal error (with $\sigma_B = 0.433$, eq. 7.16).

The current literature mass excess value [88] for 221 Fr is 13277.3(49) keV. The mass value and the corresponding mass excess (equation 3.23) obtained in this work are 221014251.45(38) µu and 13275.1(4) keV, respectively, which are in agreement

with AME 2020 data. This work improves the uncertainty by roughly an order of magnitude, from 4.9 keV to 0.36 keV.

7.3.2 Mass of ²¹⁹Rn

The first investigation and identification of ²¹⁹Rn was done in 1904 independently by French physicist A. Debierne and German chemist O. Sackur and its half life was measured to be 3.9 s [149]. The Q_{α} value of 5987.9(3) keV measured in 1962 [150] is adopted as the first contributor to the Atomic Mass Evaluation value [88]. The mass of ²¹⁹Rn given in the AME has contributions from two alpha decay channels: ²²³Ra(α)²¹⁹Rn (96.4%) and ²¹⁹Rn(α)²¹⁵Po (3.6%). As was the case with ²²¹Fr, the relative high-precision of Q_{α} , of a few hundreds of eV, does not translate into similar uncertainty in the atomic mass of ²¹⁹Rn. This uncertainty is again limited by those of the masses of the species connected via the decays.

In this work, the atomic mass of ²¹⁹Rn is measured directly for the first time. For the mass measurement of ²¹⁹Rn, three different reference ions were utilised (¹³³Cs, ⁸⁵Rb and ¹⁹⁷Au), in order to chose the reference with closest q/m value. Figure 7.9 shows the plots of the frequency ratios obtained using the PI-ICR technique for the measurements with three reference ions: ²¹⁹Rn²⁺ vs ¹³³Cs⁺, ²¹⁹Rn²⁺ vs ⁸⁵Rb⁺ and ²¹⁹Rn⁺ vs ¹⁹⁷Au⁺. The x-axis in every plot corresponds to the beginning time of the respective measurement set.

Table 7.2 details the different ion pair combinations and their respective accumulation times used for this work. It also shows the frequency ratio of the ion of interest with respect to the reference ion and their respective mass difference. It can be seen that the mass values obtained with 219 Rn²⁺ for the reference ions 133 Cs⁺ and 85 Rb⁺



Figure 7.9: Individual frequency ratios of ${}^{219}\text{Rn}^{2+}$ with respect to the reference ion a) ${}^{133}\text{Cs}^+$, (b) ${}^{85}\text{Rb}^+$ and (c) ${}^{197}\text{Au}^+$ as a function of relative time. The relative time along the x-axis has its zero at the beginning of the respective measurement sets.

are in agreement with each other as well as with the literature value from the AME 2020 [88]. However, the value from the measurements of $^{219}Rn^+$ vs $^{197}Au^+$ differs by roughly 6 sigma from the AME 2020 value and is inconsistent with the mass values obtained from the measurements $^{219}Rn^{2+}$ vs $^{133}Cs^+$ and $^{219}Rn^{2+}$ vs $^{85}Rb^+$.

Table 7.2: Reference ions and accumulation times used for the measurement of mass of ²¹⁹Rn. The table also shows the frequency ratios of the ion of interest with respect to the reference ions and the mass difference relative to the literature value from AME 2020 [88]. The number in brackets corresponds to the respective total uncertainty, including statistical and systematic errors.

Ion pair	Acc. time	Frequency ratio	$\delta R/R$	Mass diff. $[keV]$
$^{219}Rn^{2+}$ vs $^{133}Cs^+$	${\sim}800\mathrm{ms}$ & ${\sim}1\mathrm{s}$	0.8239288978(36)	4.4×10^{-9}	-0.40(44)
$^{219}Rn^{2+}$ vs $^{85}Rb^+$	$\sim \! 800 \mathrm{ms}$	1.2896312572(93)	7.2×10^{-9}	0.15(74)
$^{219}Rn^+$ vs $^{197}Au^+$	$\sim \! 1.2 \mathrm{s}$	1.1119123203(19)	$1.7{ imes}10^{-9}$	14.82(70)

The concept of the 'Birge adjustment' was implemented for the mass values (and their respective uncertainties) obtained from all the three cases and gives a Birge ratio of 13.479. The large value of the Birge ratio indicates inconsistencies in the data. Using this concept, a combined mass of $219009482.1(49) \mu u$ is obtained. However this is not considered the final mass value due to large inconsistencies and further considerations are required. The large uncertainty is due to the mass obtained with the reference ion ¹⁹⁷Au⁺, which deviates significantly from the rest of the data as shown in table 7.2. This is attributed to the large number of gold ions in the trap. The creation of ¹⁹⁷Au⁺ ions from the laser-ablation source needs high laser energy and only produces large number of ions. A count rate of roughly three ions per shot was observed at the delay line detector (at D4, figure 6.1) for ${}^{197}Au^+$ ions during measurements. Having large number of ions in the trap at the same time leads to frequency shifts [126]. The detection efficiency in normal operation is about 30%, but the presence of a large amount of noise during the measurements reduced the detector efficiency. Therefore, the issue with the large count rate was exacerbated by the increased noise. This issue was addressed only after the completion of the current measurement campaign. Detection efficiency plays a crucial role for these frequency shifts and was improved by reducing the noise on one of the channels of the delay line detector [143]. Therefore, the high count rate for ${}^{197}Au^+$ is a likely candidate to account for the shift obtained from the measurements of $^{219}Rn^+$ vs $^{197}Au^+$. The final mass of ²¹⁹Rn is therefore recalculated excluding this measurement, to provide a more precise value based on our concerns regarding the ²¹⁹Rn⁺ vs ¹⁹⁷Au⁺ dataset.



Figure 7.10: Comparison of the individual mass excess values for 219 Rn derived from decay spectroscopy experiments [150, 151]. The plot also shows the mass excess obtained from AME 2020 [88]. The black hollow circle is the value obtained using Birge method for all the three reference ions. The red circle represents the final value obtained in this work (the average of the values calculated w.r.t. the reference ions 133 Cs⁺ and 85 Rb⁺), and the shaded region is its measurement uncertainty.

The mass is obtained by averaging the values procured from measurements relative to $^{133}Cs^+$ and $^{85}Rb^+$. The Birge method (section 7.2.3) is also used in this case to estimate the uncertainty in the final mass value. Along with this, during the individual mass estimations versus the reference ions used in the final mass evaluation, the Birge ratio is used to calculate the adjusted uncertainties in each case giving the values 0.745 and 1.121, respectively, corresponding to $^{133}Cs^+$ and $^{85}Rb^+$. The mass obtained from the measurements with respect to ^{133}Cs is 219009478.25(48) µu and relative to ^{85}Rb is 219009478.52(79) µu. Therefore, this work evaluates the mass of ^{219}Rn to be 21009478.33(12) µu and the resulting mass excess is 8829.01(11) keV.

As per the AME 2020 data [88], the mass excess of 219 Rn is 8829.3(21) keV with the corresponding mass of 219009478.69(225) µu. This work improves the uncertainty by a factor of ~20 from 2.1 keV to 0.1 keV. Figure 7.10 shows the historic evolution of the mass excess from different spectroscopy experiments [150, 151], the literature value from AME 2020 [88], the values calculated in this work using the different reference ions and the final value obtained for 219 Rn.



Figure 7.11: (a) Individual frequency ratios of $^{213}\text{Bi}^{2+}$ with respect to the reference ion $^{133}\text{Cs}^+$ as a function of relative time. As in previous sections, the red dashed line represents the unadjusted value of the internal error and the black dotted line is the external error of the measurement. R_{mean} has a value of 0.8012995724(12), where the value in brackets is a combination of systematic and statistical error. (b) Comparison of the individual mass excess values for ^{213}Bi derived from decay spectroscopy experiments [145, 152, 153, 154, 155]. The plot also shows the mass excess obtained from AME 2020 [88]. The right-most value is obtained in this work, and the shaded region is its measurement uncertainty.

7.3.3 Mass of ²¹³Bi

The first experiment reported in Atomic Mass Evaluation contributing to ²¹³Bi was performed by A. A. Vorobev *et al.* in 1960 [152] and it obtained a Q_{α} value of 7200.3(30) keV. The two dominant contributors in AME 2020 [88] towards the mass evaluation of ²¹³Bi are the decays ²¹⁷At(α)²¹³Bi (76.7%) and ²¹³Bi(β^{-})²¹³Po (23.3%). Even though decay experiments have been performed over a span of several years, the uncertainty of the Q_{α} value is about half of the current uncertainty of the literature mass excess value (AME 2020 [88]). The high mass uncertainty is again due to the large atomic mass uncertainty of the elements linked through the decay chains (²¹⁷At, ²¹³Po).

The cyclotron frequency of ²¹³Bi²⁺ ions was measured relative to the reference ion ¹³³Cs⁺ using PI-ICR to obtain a mean frequency ratio of $R_{mean} = 0.8012995724(12)$ with a relative uncertainty $\delta R/R = 1.6 \times 10^{-9}$. Accumulation times of ~500ms and ~1200 ms were used for the measurement. In figure 7.11a the frequency ratios are plotted vs relative time, with the *x*-axis zero as beginning time of the measurement set. This work's resulting atomic mass and mass excess values are 212994381.67(17) µu and -5233.4(2) keV, respectively, which are in good agreement with the literature value from AME 2020 [88]. The concept of 'Birge adjustment' is applied (section 7.2.3) giving $\sigma_B = 0.249$. The current work improves the uncertainty by a factor of ~25 as compared to the AME 2020 value. Figure 7.11b shows the mass excess values obtained from multiple decay spectroscopy experiments [145, 152, 153, 154, 155, 156] and the literature value of mass excess from AME 2020 [88].

7.3.4 Mass of ²¹¹Pb

The decay spectroscopy experiment performed by C. R. Cothern and R. D. Connor in 1965 was the first experiment included in Atomic Mass Evaluation concerning the mass measurement of ²¹¹Pb [157]. As per the AME 2020 [88], ²¹⁵Po(α)²¹¹Pb (95.8%) and ²¹¹Pb(β^-)²¹¹Bi (4.2%) are the main contributors towards the mass evaluation of ²¹¹Pb. The Q_{α} value of 7526.5(8) keV, given by the group of Grennberg *et al.* [151], has an uncertainty of 0.8 keV which is lower than the current mass uncertainty of ²¹¹Pb by a factor of 3. As already discussed in the previous sections, this is because the atomic mass of ²¹¹Pb is limited by the imprecise knowledge of the atomic mass value of the parent element ²¹⁵Po (or daughter ²¹¹Bi in the beta decay).

The obtained frequency ratios of ${}^{211}\text{Pb}^{2+}$ with respect to the reference ion ${}^{133}\text{Cs}^+$ are shown in figure 7.12a. All the measurements use an accumulation time of $\sim 1200 \text{ ms}$



Figure 7.12: (a) Individual frequency ratios of $^{211}\text{Pb}^{2+}$ with respect to the reference ion $^{133}\text{Cs}^+$ as a function of relative time. As before, the red dashed line represents the internal error before Birge adjustment and the black dotted line is the external error of the measurement. R_{mean} has a value of 0.7937541548(08). (b) Comparison of the individual mass excess values for ^{211}Pb derived from decay spectroscopy experiments [151, 157]. The plot also shows the mass excess obtained from AME 2020 [88]. The right-most value is obtained in this work, and the shaded region is its measurement uncertainty.

and employ the PI-ICR technique (section 3.6). A mean frequency ratio of $R_{mean} = 0.7937541548(08)$ is obtained in this work. This corresponds to a relative uncertainty of $\delta R/R = 1.1 \times 10^{-9}$, after including a Birge adjustment of $\sigma_B = 0.377$ in the estimation of the internal error (section 7.2.3). The internal error before the adjustment and the external error are shown in figure 7.12a. The mass value and the corresponding mass excess (equation 3.23) obtained in this work are 210988735.65(11) µu and -10492.7(1) keV, respectively. This is in agreement with the AME 2020 mass excess value of -10493.0(22) keV [88]. This work improves the uncertainty in mass excess by a factor of 20, from 2.2 keV to 0.1 keV. Historic progression of various mass excess values obtained from the decay spectroscopy experiments [151, 157] and the value listed in AME 2020 [88], together with the mass excess determined in this work, are shown in figure 7.12b.

7.3.5 Mass of ²⁰⁹Pb

The first experiment involving ²⁰⁹Pb was carried out by A. Sperduto and W. W. Buechner at M.I.T., and was published in 1964 [158]. As per the Atomic Mass Evaluation [88], the three biggest contributors towards the mass measurement of ²⁰⁹Pb are: ²⁰⁹Pb(β^{-})²⁰⁹Bi (86.9%), ²⁰⁸Pb(d, p)²⁰⁹Pb (11.1%) and ²¹³Pb(α)²⁰⁹Pb (2%). Using the equations presented in section 3.7, the mass excess resulting from various decay spectroscopy experiments [153, 155, 158, 159, 160, 161, 162, 163] can be obtained from the decay energies. The masses of the corresponding elements have been taken from the most recent AME [88]. Currently, the atomic mass excess of ²⁰⁹Pb is evaluated in AME 2020 to be -17614.57(174) keV [88].

The cyclotron frequency of ${}^{209}\text{Pb}^{2+}$ is measured with respect to the reference ion ${}^{133}\text{Cs}^+$ and the obtained frequency ratios are shown in figure 7.13a. The measurements are performed for an accumulation time of ~1200 ms. This gives a mean frequency ratio of $R_{mean} = 0.7862012402(13)$ and a corresponding relative uncertainty of $\delta R/R = 1.6 \times 10^{-9}$. In order to estimate the final uncertainty, a Birge ratio of $\sigma_B = 1.023$ is included in the internal error.

Even though the decay chain of 225 Ac (figure 7.1) shows that 209 Pb should coexist with its isobar 209 Tl, only the 209 Pb ions are observed distinctly at the detector. There are a few indications of the presence of 209 Tl upon using magnetron splitting (section 3.6.2) between species, but due to the strong background, it was difficult to unequivocally identify the isobar. The obtained mass excess from this work is -17608.13(16) keV corresponding to the mass 208981096.90(17) µu. This work improves the uncertainty from 1.76 keV to 0.16 keV, almost an order of magnitude.



Figure 7.13: (a) Individual frequency ratios of ${}^{209}\text{Pb}^{2+}$ with respect to the reference ion ${}^{133}\text{Cs}^+$ as a function of relative time. The red dashed line represents the internal error and the black dotted line is the external error of the measurement. R_{mean} has a value of 0.7862012402(13). (b) Comparison of the individual mass excess values for ${}^{209}\text{Pb}$ derived from decay spectroscopy experiments [153, 155, 158, 159, 160, 161, 162, 163]. The plot also shows the mass excess obtained from AME 2020 [88]. The right-most value is obtained in this work, and the shaded region is its measurement uncertainty.

Figure 7.13b shows the mass excess obtained in this work together with the historic evolution and comparison of various mass excess values obtained from the decay spectroscopy experiments and the literature value listed in AME 2020. As can be seen from the figure, the mass excess of 209 Pb is not in agreement with the AME 2020 mass excess value of -17614.57(175) keV but it agrees with the most recent experimental value from the measurements performed using the reaction 48 Ca + 243 Am with the decay station TASISpec at TASCA, GSI by A. Saamark-Roth *et al.* [163].

7.3.6 The case of the mass doublet : ²⁰⁷Tl and ²⁰⁷Pb

The mass doublet of ²⁰⁷Tl and ²⁰⁷Pb has a mass difference of ~1.4 MeV. As depicted in the decay chain of ²²³Ra (figure 7.1), ²⁰⁷Tl decays to ²⁰⁷Pb by β^- decay. During the mass measurement of either of the elements of this doublet using the PI-ICR technique (section 3.6), the other isobaric species is also present in the trap. The count rate was so low that there were never two ions in the trap at the same time, within the limits given by the detector efficiency. On the detector, two different spots are seen for the cyclotron mode and, with a particular setting of t_s , an angle difference can be induced between the two ion spots in the magnetron mode as well (magnetron splitting, section 3.6.2). In the present work, various measurements were taken for



Figure 7.14: Scheme depicting the classification of different data sets accumulated for the mass measurements of 207 Tl.

different accumulation times (~500 ms and ~1200 ms). The measurements can be categorised in different sets based on a) choice of the reference ion, b) isobar used to determine conversion frequency ν_c and c) whether the magnetron splitting was used.

As a brief overview of the historic progression and atomic mass evaluation of ^{207}Tl and ^{207}Pb , the first experiment to measure the α -decay energy of ^{207}Tl using $^{211}\text{Bi}(\alpha)^{207}\text{Tl}$ was performed in 1961 by A. Rytz [164]. The obtained Q_{α} value of 6749.5(7) keV is the first reported contribution towards determination of the ^{207}Tl mass in Atomic Mass Evaluation [88]. The uncertainty in the measurement of the Q_{α} value is much smaller than the reported mass uncertainty of ~6 keV. Along with the α -decay of ^{211}Bi , which contributes significantly (42.4%) towards the mass evaluation of ^{207}Tl , AME 2020 [88] lists two more contributors $^{207}\text{Tl}(\beta^-)^{207}\text{Pb}$ (44.9%) and $^{205}\text{Tl}(t, p)^{207}\text{Tl}$ (12.8%) to the masses.

Furthermore, the first reported experiment to measure the nuclear structure and properties of ²⁰⁷Pb is given in [165]. The β -decay experiment was performed by the group of W. F. Davidson *et al.* and gives an endpoint energy of 1431(8) keV. The β -decay ²⁰⁷Tl(β^{-})²⁰⁷Pb contributes only minimally (0.7%) towards the atomic mass evaluation of ²⁰⁷Pb. As per the AME 2020 [88], there are two major contributors, ²⁰⁶Pb(n, γ)²⁰⁷Pb (86.3%) and ²⁰⁷Pb(n, γ)²⁰⁸Pb (13%), which influence the mass evaluation of ²⁰⁷Pb. The uncertainty in the decay energy values of the (n, γ) reaction is of the order of a few hundred electronvolts. Over the span of last 50 years, various experiments have been performed but the uncertainty in the mass of ²⁰⁷Tl and ²⁰⁷Pb did not change significantly. The large value of mass uncertainty compared to the



Figure 7.15: Mass excess values for different measurement sets of (a) ${}^{207}\text{Tl}^{2+}$ and (b) ${}^{207}\text{Pb}^{2+}$. ${}^{133}\text{Cs}^+, \nu_{Pb,s}$ denotes the measurements done with the reference ion source as ${}^{133}\text{Cs}^+$ using the ν_c of ${}^{207}\text{Pb}$ and with magnetron splitting. The measurements for each of the isobaric species are performed using two different reference ions, at two excitation frequencies, with and without magnetron splitting. The difference of the mass excess value from all the eight measurement files for each isobar are shown with their respective uncertainties. The red band shows the uncertainty in AME 2020 value [88].

achieved experimental uncertainties in the decay energies can again be attributed to the mass uncertainty of the species involved in the contributing decay chains. The mass excess can be obtained from the decay energies by following the procedure shown in 3.7. The masses of the corresponding species have been taken from the most recent Atomic Mass Evaluation [88]. This work presents the first direct mass measurements of ²⁰⁷Tl and ²⁰⁷Pb using the PI-ICR technique.

For the mass evaluation, the data set measurement and analysis for both of the isobaric species are carried out in a similar manner. The mass measurement process for 207 Tl has been discussed first. A similar process is followed for the mass measurement of 207 Pb. As depicted in figure 7.14, the analysis concerning the mass of 207 Tl has been divided into two major sets. In the first case, the 207 Tl²⁺ frequency ratio is calculated relative to the reference ion 133 Cs⁺. However in the second case, the ion frequency ratio of 207 Tl²⁺ is measured with respect to 207 Pb²⁺, which is measured at the same instant as the ion of interest (207 Tl²⁺). The measurements performed using the isobar 207 Pb²⁺ are insensitive to some of the systematic uncertainties presented in section 7.2.4. Since in this case both of the reference ion species are measured at the same time, the temporal changes in magnetic field are irrelevant. This eliminates the need for interpolation of the reference ion frequency. Additionally, the masses

²⁰⁷ Tl		²⁰⁷ Pb		
Ion pair	m (µu)	Ion pair	m (µu)	
$^{133}\mathrm{Cs}^+, \nu_{Pb,s}$	206977418.64(39)	$^{133}\mathrm{Cs}^+, \nu_{Pb,s}$	206975900.65(34)	
$^{133}\mathrm{Cs}^+, \nu_{Pb,n}$	206977424.08(16)	$^{133}\mathrm{Cs}^+, \nu_{Pb,n}$	206975896.53(33)	
$^{133}\mathrm{Cs}^+, \nu_{Tl,s}$	206977421.83(37)	$^{133}\mathrm{Cs}^+, \nu_{Tl,s}$	206975902.50(40)	
$^{133}\mathrm{Cs}^+, \nu_{Tl,n}$	206977422.95(22)	$^{133}\mathrm{Cs}^+, \nu_{Tl,n}$	206975900.07(53)	
$^{207}\mathrm{Pb}^+, \nu_{Pb,s}$	206977415.06(144)	$^{207}\mathrm{Tl}^+, \nu_{Pb,s}$	206975899.36(587)	
$^{207}\mathrm{Pb}^+, \nu_{Pb,n}$	206977424.44(134)	$^{207}\mathrm{Tl}^+, \nu_{Pb,n}$	206975891.00(586)	
$^{207}\mathrm{Pb}^{+}, \nu_{Tl,s}$	206977416.47(153)	$^{207}\mathrm{Tl}^+, \nu_{Tl,s}$	206975899.09(591)	
$^{207}\mathrm{Pb}^+, \nu_{Tl,n}$	206977419.84(164)	$^{207}\mathrm{Tl}^+, \nu_{Tl,n}$	206975895.66(594)	

Table 7.3: Calculated masses of ²⁰⁷Tl and ²⁰⁷Pb for different measurement sets.

of the isobars differ only by ~1.4 MeV, due to which the mass dependent systematic uncertainty is $R_{1,mean} \approx 10^{-14}$ (section 7.2.4), so it does not play a role within the achieved statistical uncertainties.

For each of the cases described above, there are two further sub-divisions based on the frequency used for the quadrupolar excitation. For the isobars, which do not differ significantly in mass and are measured in the same data file, either of the masses can be used to get an approximation for the excitation frequency ν_c . Hence, the measurement sets are recorded with the quadrupolar excitation of both isobars: ²⁰⁷Tl and ²⁰⁷Pb.

In addition to this, the concept of magnetron splitting (section 3.6) was included in the measurements in an attempt to avoid the so-called 'Kretzschmar systematic' [166, 167, 168]. This systematic effect arises when the driving frequency does not match the cyclotron frequency. In the case of mass measurements of isobars or relatively high energy isomers using the conventional PI-ICR technique, the cyclotron image has different spots corresponding to each of the isomers/isobars. However, because of the mass independence of the magnetron motion, a single magnetron spot is obtained. In such a scenario, the effects due to off-resonant excitation are not the same for the cyclotron and magnetron spot thereby introducing additional systematic effects. In an attempt to avoid this systematic effect, a splitting was introduced in the magnetron image (magnetron splitting) by changing the time t_s (figure 3.9), where each of the spots observed in the magnetron image corresponds to an isobaric ion species.

Figure 7.15 represents the the mass excess values for all the measured data sets and the corresponding uncertainties observed for both of the isobars in comparison to the AME 2020 values. Here, ${}^{133}Cs^+$, $\nu_{Pb,s}$ denotes the measurements done with the



Figure 7.16: Individual frequency ratios obtained using the PI-ICR technique with respect to the reference ion ${}^{133}\text{Cs}^+$ for (a) ${}^{207}\text{Tl}^{2+}$ and (b) ${}^{207}\text{Pb}^{2+}$ as a function of relative time. The data set depicted here corresponds to the measurements performed using the excitation frequency of the ion of interest and by applying magnetron splitting. As in the previous cases, the red dashed line represents the uncorrected internal error and the black dotted line is the external error of the measurement. The R_{mean} for ${}^{207}\text{Tl}^{2+}$ has a value of 0.7786632374(28) and ${}^{207}\text{Pb}^{2+}$ has $R_{mean} = 0.7786575146(26)$ in (b), where the value in brackets is a combination of systematic and statistical error.

reference ion source as ¹³³Cs⁺ using the ν_c of ²⁰⁷Pb and with magnetron splitting. Likewise, the term ²⁰⁷Pb²⁺, $\nu_{Tl,n}$ denotes the measurements done with the reference ion source as ²⁰⁷Pb²⁺ using the ν_c of ²⁰⁷Tl and with *no* magnetron splitting. The value of the masses obtained from these cases have been depicted in table 7.3. Using the Birge method, these mass values are combined to obtain a mass of 206977422.99(66) for ²⁰⁷Tl with a Birge ratio of 5.817. For ²⁰⁷Pb, mass value of 206975899.62(89) is obtained and the corresponding Birge ratio is 4.662. The Birge ratios for both of the doublet species are significantly greater than 1, indicating inconsistencies in the data. It should be noted that these mass values given above for ²⁰⁷Pb/²⁰⁷Tl are not the final result from this work but are listed just for comparison with the final value specified later in this section.

From figure 7.15 and table 7.3, it can be seen that the deviation between magnetron splitting and no splitting is very pronounced even when the ion that is being measured is excited using its own ν_c . This points towards the presence of significant 'Kretzschmar systematic effects' and discourages the use of data without magnetron splitting. It should also be noted that when the isobaric species are used as reference, the conversion pulse for the reference ion is off-resonance. Since the measurements are made at the same time for both of the ions, in any such measurement, the quadrupo-



Figure 7.17: (a) Comparison of the individual mass excess values for ²⁰⁷Tl derived from decay spectroscopy experiments [151, 164, 165, 169] along with the mass excess obtained from AME 2020 [88] and the present work.(b) Comparison of the individual mass excess values for ²⁰⁷Pb derived from decay spectroscopy experiments [165, 170, 171, 172, 173, 174] along with the mass excess obtained from AME 2020 [88] and the present work. The red circle corresponds to the value obtained in this work, and the shaded region is its measurement uncertainty. The value corresponding to 'all data' is depicted for comparison but not considered as the final result.

lar excitation frequency can correspond to only one of the ions. This puts the other species 'off-resonance', hence shifting that ion's measured frequency. As an example, if ²⁰⁷Pb is chosen as the reference ion for the ²⁰⁷Tl measurement, with the excitation set at ²⁰⁷Tl's cyclotron frequency, the reference ion will be driven off-resonance and hence influenced by the Kretzschmar systematic.

In the future it would be possible to measure the isobars first with one frequency and then with the other, in both cases with magnetron splitting. For the present work, the measurement sets are chosen relative to the reference ion $^{133}Cs^+$, with the excitation cyclotron frequency corresponding to the ion of interest along with magnetron splitting of the spots. This corresponds to $^{133}Cs^+$, $\nu_{Tl,s}$ for evaluation of the ^{207}Tl mass and to $^{133}Cs^+$, $\nu_{Pb,s}$ for the ^{207}Pb mass measurement.

The frequency ratios obtained from both of these measurement sets have been represented as a function of relative time in figure 7.16.

The reported literature uncertainty in the mass of 207 Tl in AME 2020 [88] is 5.4 keV. This work reduces the uncertainty by about an order of magnitude to 0.34 keV. For the mass evaluation of 207 Pb the uncertainty in the literature value of AME 2020 is reduced by a factor of ~20: from 5.55 keV [88] to ~0.32 keV. Figure

Ion pair	Frequency ratio (R_{mean})	$\delta R/R$	Obtained mass (µu)
$^{207}\text{Tl}^{2+}$ vs $^{133}\text{Cs}^+$	0.7786632374(27)	3.5×10^{-9}	206977421.83(37)
$^{207}\text{Pb}^{2+} \text{ vs} {}^{133}\text{Cs}^{+}$	0.7786575146(26)	3.3×10^{-9}	206975900.65(34)

Table 7.4: Mean frequency ratio, relative uncertainty and obtained mass of 207 Tl and 207 Pb using magnetron splitting relative to the reference ion 133 Cs⁺.

7.17a shows the historic progression and comparison of various mass excess values obtained from the decay spectroscopy experiments [151, 164, 165, 169] for ²⁰⁷Tl, the literature value from AME 2020 [88], the average value of all the cases (depicted in figure 7.14) using the Birge method and the final value obtained in the present work. A similar comparison for ²⁰⁷Pb with the decay spectroscopy experiments [165, 170, 171, 172, 173, 174] is shown in figure 7.17b. The measured values of the frequency ratios, relative uncertainties $\delta R/R$ and masses of both these ions have been shown in table 7.4.

Recently, a direct mass measurement of ²⁰⁸Pb has been reported by K. Kromer *et al.* [175]. The experiment was performed at the PENTATRAP facility in Heidelberg, Germany, by measuring the cyclotron frequency ratio for ²⁰⁸Pb⁴¹⁺ ions with respect to ¹³²Xe²⁶⁺ ions using the Fourier-transform ion-cyclotron-resonance-technique [176]. As discussed in the paper, the new mass value for ²⁰⁸Pb also changes the currently known masses and uncertainties (as listed in AME 2020 [88]) for several other elements including ²⁰⁷Pb. The improved mass value for ²⁰⁷Pb is given as 206975895.39(06) µu and its uncertainty is reported to be improved by a factor of ~21. This has also been depicted in figure 7.17b. The new AME value for ²⁰⁷Pb is more precise than the mass value obtained in this work. Furthermore, the mass obtained in the current work differs by ~15 σ from the new AME value for ²⁰⁷Pb [175] differs significantly from the AME 2020 mass value. Since the experimentally observed values are not in agreement with other, this discrepancy requires further studies.

7.4 Summary of the measured atomic masses

An overview of the difference between the mass values of the species measured in this work (namely ²⁰⁷Tl, ²⁰⁷Pb, ²⁰⁹Pb, ²¹¹Pb, ²¹³Bi, ²¹⁹Rn and ²²¹Fr) and of the Atomic Mass Evaluation 2020 [88] has been shown in figure 7.18. The black data points represent the mass values and uncertainties of these species from AME 2020 and the red band shows the uncertainties of the evaluations from this work. As can be seen

from figure 7.18, the obtained final values for masses of ²⁰⁷Tl, ²¹¹Pb, ²¹³Bi, ²¹⁹Rn, and ²²¹Fr agree well within the uncertainty from AME 2020. This work improves the mass uncertainties given in AME 2020 by at least an order of magnitude. The mass of ²⁰⁹Pb agrees with the value obtained using Q_{α} for ²¹³Pb(α)²⁰⁹Pb from the TASCA experiment at GSI [163], but does not agree with the literature value from AME 2020.

The new AME mass value for ²⁰⁷Pb from the PENTATRAP experiment [175] is given as 206975895.39(06) µu. This value is ~6 times more precise than the value obtained in this work. The mass value for ²⁰⁷Pb obtained in this work differs from the AME 2020 and new PENTATRAP based AME value. Further studies should be carried out to understand this discrepancy. Based on the masses obtained in this work, an evaluation to improve the linked (directly and indirectly) masses through the Atomic Mass Evaluation is in progress. The mass of ²⁰⁹Bi is linked to ²⁰⁹Pb through the beta decay ²⁰⁹Pb(β^{-})²⁰⁹Bi and an improvement in the mass of ²⁰⁹Pb (and other elements in the decay chain as seen in figure 7.1) can directly affect the current knowledge of ²⁰⁹Bi's mass. Similarly, the masses of ²⁰⁵Pb and ²⁰⁵Tl doublet are linked to the masses of elements evaluated during this work: ²⁰⁵Pb is linked to



Figure 7.18: Mass difference between AME 2020 [88] data and evaluations from this work along with their uncertainties. The black squares are the final results from AME 2020 and the red band represents the uncertainty values from this work.

²⁰⁷Pb by a series of (n,γ) reactions and ²⁰⁵Tl to ²⁰⁷Tl by (t,p) reaction. The ²⁰⁵Pb / ²⁰⁵Tl doublet is of great importance for the nuclear astrophysics as an s-process cosmochronometer [22] and also for the LOREX project (LORandite EXperiment) to determine the nuclear matrix elements for the neutrino capture of ²⁰⁵Pb and bound-state beta decay of ²⁰⁵Tl⁸¹⁺ [23, 24].

The evaluated atomic masses of the elements obtained in this work from the recoilion sources have been summarised in table 7.5 and figure 7.18. The table shows mass excess values and relative uncertainties $(\delta m/m)$ evaluated in this work and mass excess values from AME 2020 [88].

Table 7.5: The mass excess values uncertainty and obtained mass excess of various elements compared to the AME 2020 [88] values are shown in the table. The masses are measured using PI-ICR relative to a well-known reference ion.

Element	ME_{AME} [keV]	$ME_{Thiswork}$ [keV]	$m_{Thiswork}$ [µu]	$(\delta m/m)$
221 Fr	13277.25(488)	13275.14(36)	221014251.45(38)	1.7×10^{-9}
219 Rn	8829.34(210)	8829.01(11)	219009478.33(12)	$5.5{\times}10^{-10}$
²¹³ Bi	-5231.67(508)	-5233.45(15)	212994381.67(17)	7.8×10^{-10}
$^{211}\mathrm{Pb}$	-10493.01(226)	-10492.67(10)	210988735.65(11)	5.4×10^{-10}
²⁰⁹ Pb	-17614.57(174)	-17608.13(16)	208981096.90(17)	8.0×10^{-10}
²⁰⁷ Pb	-22451.96(115)	-22448.40(32)	206975900.65(34)	1.7×10^{-9}
²⁰⁷ Tl	-21034.44(544)	-21031.44(34)	206977421.83(37)	1.8×10^{-9}

Chapter 8 Conclusion and Outlook

Within the scope of this work, non-destructive electronic detection systems were fabricated and experimental studies were conducted on the ion ensemble cooling and cleaning schemes to obtain a pure Ar^{13+} cloud for the high-precision *g*-factor measurements at the ARTEMIS Penning trap experiment. In addition to this, high-precision mass measurements of the decay products from the recoil-ion sources ²²⁵Ac and ²²³Ra were performed at the SHIPTRAP mass spectrometer.

The measurements on ion clouds in the ARTEMIS experiment described in this work have been carried out during the two measurement campaigns of 2019 and 2021. Between these two measurement campaigns, the necessary updates were performed on the non-destructive detection systems as discussed in section 4.4. Three detection systems are currently connected to the ARTEMIS Penning trap: an axial resonator at the creation trap and an axial resonator and a cyclotron resonator at the spectroscopy trap were fabricated using a normal conducting coil within the scope of this work.

In addition to these the cryogenic amplifiers have been fabricated with the new rectangular printed circuit boards which are now mounted on the body of the resonator. The change in the orientation of drain-to-source current of the transistor in the amplifier board relative to the magnetic field axis has improved the sustainability of the boards tremendously. Furthermore, the components and techniques that are used for the fabrication have been carefully chosen to work in the cryogenic, high magnetic field environment of the trap. During the course of this work, efforts have already been started to use the resonators with superconducting coil and superconducting housing to produce resonators with very high quality factors. These housings and a helical coil have already been prepared. A toroidal resonator with superconducting coil and copper housing having a quality factor of 4000 at 2.5 MHz has been tested and developments are ongoing to include more resonators with toroidal geom-

etry in the trap. Furthermore, upgrades to the cryogenic amplifier boards based on the knowledge from this work are also foreseen in the near future.

The knowledge of the load capacitance from the trap as seen by the resonator is necessary in order to determine the center frequency and number of turns for the fabrication of resonator coils. As discussed in section 5.1, the effect of the load trap capacitance is much more pronounced for the cyclotron resonator connected to the spectroscopy trap (CRES ST) due to the variable capacitor (varactor diode board) connected to it. Detailed studies regarding the change in the center frequency, quality factor and signal-to-noise ratio have been performed for different bias voltages of the varactor diode and have been listed in section 4.4.

During the measurement campaigns of 2019 and 2021, the ion creation, cooling and cleaning techniques for boron-like argon, Ar^{13+} have been studied in detail and the results have been reported in chapter 5. The creation parameters for the Ar^{13+} ion cloud a re optimised. The cloud is successfully transported between the two traps of the ARTEMIS set-up: the creation trap and the spectroscopy trap. The transport of the hot cloud between the traps is used to cool the ions by scraping off the hotter ions along with resistive cooling, which is an eminent feature of the scan of the trapping potential. This combined procedure of transport and resistive cooling is observed to be faster than the conventional resistive cooling technique. A study of the area under the curve and relative frequency shift of a particular charge state with successive transports is reported in this work. However, more extensive studies should be performed in the future focused on this cooling technique to obtain a detailed analysis for the ion number loss and final energy of the ions.

A pure cloud of Ar¹³⁺ ions is obtained by using the Stored Waveform Inverse Fourier Technique (SWIFT). For the first time, this selective cleaning process has been applied to the ions stored in the spectroscopy trap. A comparative analysis has been done for the implementation of SWIFT on hot and cooled ion clouds trapped in the creation trap and the spectroscopy trap. Based on the area under the curve in the potential scan, it has been observed that the most effective configuration for cleaning the ion cloud is on the cooled ions stored in the creation trap. However, the combination of transport of a cooled cloud and implementation of SWIFT in the spectroscopy trap has not been studied in detail within the scope of this work. This can be performed in the future in order to comprehend fully the applicability of the cleaning technique in the spectroscopy trap. This is specifically crucial for the ions that are injected through external ion sources such as EBITs and through the HITRAP facility. Due to the successful transport, cleaning and storage of ions in the spectroscopy trap, estimation of the residual gas pressure is performed by measuring the storage time of the ions. For Ar^{13+} , the storage time in the spectroscopy trap is ~18 hours. Such measurements in the spectroscopy trap using highly charged argon ions have been reported for the first time and a residual gas pressure of better than 3.1×10^{-14} mbar is observed in the trap. In the future, efforts should be made to improve the pressure inside the trap in order to facilitate longer storage times for highly charged ions like Bi⁸²⁺.

The connection of the ARTEMIS experiment to the HITRAP beamline has already been established and the details regarding this set-up can be found in the upcoming thesis from J. Klimes [97]. Preliminary tests from these beamline developments are currently ongoing. Therefore, the first proof of concept measurements for the lasermicrowave double-resonance spectroscopy at the ARTEMIS Penning trap experiment are foreseen in the near future.

In the last quarter of 2021, high-precision mass measurements were carried out at the SHIPTRAP experimental set-up. The measurements were performed on the radioactive decay products of the recoil-ion sources ²²⁵Ac and ²²³Ra installed in the cryogenic gas cell at SHIPTRAP. During the measurement campaign, the masses of ²²¹Fr, ²¹³Bi and ²⁰⁹Pb from the decay chain of ²²⁵Ac, and ²¹⁹Rn, ²¹¹Pb, ²⁰⁷Tl and ²⁰⁷Pb from the decay chain of ²²³Ra were measured using the Phase-Imaging Ion-Cyclotron-Resonance technique (PI-ICR). This work presents the first-ever direct mass measurements for these radio-nuclides.

The measured masses are in agreement with the literature masses from AME 2020 [88] except for ²⁰⁷Pb and ²⁰⁹Pb. However, the obtained mass of ²⁰⁹Pb agrees with the recent experiment performed at the TASCA experiment at GSI, as described in section 7.3.5. The absolute mass uncertainties are improved by at least an order of magnitude in comparison to the literature values from the Atomic Mass Evaluation 2020 and are now in the range of a few hundreds of eV/c^2 . Within this work, relative mass uncertainties of $\delta m/m \sim 10^{-9}$ are achieved. Currently there are no nuclides in this region of the nuclear chart with a relative precision $\delta m/m > 10^{-9}$. High-precision mass measurements in this domain are important since they provide new anchor points to determine the atomic masses of heavier nuclei.

Additionally, the mass of ²⁰⁹Pb is linked to ²⁰⁹Bi through beta decay, ²⁰⁹Pb(β^{-})²⁰⁹Bi. The mass precision of ²⁰⁹Bi is expected to improve after including the high-precision masses from this work in the Atomic Mass Evaluation. ²⁰⁹Bi is the first candidate towards the bound electron *g*-factor measurement in heavy, highly charged ions at

ARTEMIS. However, a direct mass measurement for ²⁰⁹Bi can be performed in the future using the SHIPTRAP experimental set-up, since ²⁰⁹Bi is commercially available in the form of a thin foil. The laser-ablation technique can be utilised to produce singly charged ²⁰⁹Bi ions. For such a measurement, the reference ion species can be chosen from the nuclides measured during this work in replacement of the commonly used ¹³³Cs⁺ to avoid the mass-dependent systematic shifts. This is possible because the present work improves the precision of masses in this domain of the nuclear chart. In the light of recent update in the mass of ²⁰⁷Pb through the direct mass measurement of ²⁰⁸Pb at the PENTATRAP experiment and the discrepancies with the mass value obtained in this work, further studies on the mass of ²⁰⁷Pb should be carried forward.

The masses of ²⁰⁵Pb and ²⁰⁵Tl doublet are also linked to the masses of radionuclides evaluated during this work. This doublet is of great importance for the nuclear astrophysics as an slow-neutron capture process cosmochronometer and also for the LOREX project (LORandite EXperiment) to determine the nuclear matrix elements related to the doublet, as detailed in section 7.4. An evaluation based on the results obtained in this work to comprehend the effect on the linked masses through the Atomic Mass Evaluation is in progress.

There have been various mass measurements for the heavy and exotic nuclei at the SHIPTRAP set-up with the most recent ones being 251 No, 254,256 Lr and 257 Rf having a mass-resolving power of $\delta m/m \sim 10^{-9}$ [87]. The attempts towards the mass measurements for 257,258 Db would be addressed in the experimental campaign in 2024. In parallel to these, a second dedicated Penning trap set-up (SHIPTRAP-2) is under development and would be integrated in the current beamline. This setup is devoted to the implementation of the non-destructive Fourier-Transform Ion-Cyclotron Measurement Technique (FT-ICR) for the studies of heavier and more exotic elements with reduced production rates.

Appendix A

The cryogenic test-stand for ARTEMIS

In order to test the components of non-destructive detection system described in section 4.4 at cryogenic temperature, an external test-stand is used. The test-stand is located on top of the HITRAP platform in GSI, Darmstadt. The detection components such as resonators and cryogenic amplifier boards are tested in the test stand at a temperature of 4K prior to installation in the ARTEMIS set-up. A two stage pulsed cryocooler SRP-082B2S along with a F-70S liquid helium compressor from SUMITOMO cryogenics, identical to the cryocooler assembly in the ARTEMIS Penning trap assembly, is used to attain cryogenic temperatures. The test-stand provides the conditions similar to the magnet bore of ARTEMIS (without the magnetic field) where the resonators are installed on the 4-leg structure. The frequency spectrum of the resonators mounted in the test stand is observed using E5080AENA Network analyser from KEYSIGHT. Figure A.1 shows the test stand assembly with the vacuum pumps and two stage cryocooler. Details on the test-stand set-up and wiring plan can be found in [177].

Appendix A. The cryogenic test-stand for ARTEMIS



Figure A.1: The cryogenic test-stand assembly for ARTEMIS.

Appendix B

Characterisation of varactor diode board at ARTEMIS

Table B.1: Characterisation of varactor diode board. The heaters of the varactor board are set to 2 V during these measurements. The capacitance values shown here correspond to the entire system except the resonator, i.e. the load capacitance for the cyclotron resonator at 4 K. SNR corresponds to the signal-to-noise ratio of the frequency spectrum at the resonance frequency. The frequency depicted here is the central frequency of frequency spectrum as seen on the KEYSIGHT N9000B spectrum analyser.

Bias voltage [V]	Frequency [MHz]	Q-factor	SNR [dB]	Capacitance [pF]
0.0	33.58	51.2	32.7	33.3
0.5	34.43	63.2	34.6	31.5
1.0	34.45	71.3	35.6	31.4
1.5	34.97	82.9	37.2	30.4
2.0	35.35	91.1	37.9	29.7
2.5	35.83	99.2	39.0	28.8
3.0	35.92	96.6	39.1	28.6
3.5	36.10	107.8	39.1	28.3
4.0	36.22	108.8	40.1	28.1
4.5	36.36	107.2	40.3	27.8
5.0	36.43	107.5	40.3	27.7
5.5	36.54	107.5	40.4	27.5
6.0	36.62	102.9	41.5	27.4
6.5	36.67	108.2	40.4	27.3
7.0	36.74	118.1	40.6	27.2
7.5	36.76	117.8	40.5	27.1
8.0	36.81	110.9	40.8	27.0
8.5	36.85	111.3	41.1	27.0
9.0	36.89	111.1	41.2	26.9
9.5	36.91	111.2	41.2	26.9
10	36.94	111.3	41.2	26.8

References

- R. P. Feynman. Space-Time Approach to Quantum Electrodynamics. *Phys. Rev.*, 76:769–789, Sep 1949. doi:10.1103/PhysRev.76.769.
- [2] David J. Griffiths. Introduction to Elemantary Particle Physics. John Willey and Sons, 1987. ISBN 0-471-60386-4.
- [3] David J. Gross et al. Ultraviolet Behavior of Non-Abelian Gauge Theories. *Phys. Rev. Lett.*, 30:1343–1346, Jun 1973. doi:10.1103/PhysRevLett.30.1343.
- [4] H. David Politzer. Reliable Perturbative Results for Strong Interactions? *Phys. Rev. Lett.*, 30:1346–1349, Jun 1973. doi:10.1103/PhysRevLett.30.1346.
- [5] A. Salam et al. Electromagnetic and weak interactions. *Physics Letters*, 13(2):168–171, 1964. ISSN 0031-9163. doi:https://doi.org/10.1016/0031-9163(64)90711-5.
- [6] A. Einstein. The Foundation of the General Theory of Relativity. Annalen der Physik, 49:769–822, 1916. ISSN 0031-9163. doi:http://dx.doi.org/10.1002/andp.19163540702.
- [7] X. Fan et al. Measurement of the Electron Magnetic Moment. *Phys. Rev. Lett.*, Sept. 2022. doi:10.48550/arXiv.2209.13084.
- [8] D. Hanneke et al. New measurement of the electron magnetic moment and the fine structure constant. *Phys. Rev. Lett.*, 100:120801, Mar 2008. doi:10.1103/PhysRevLett.100.120801.
- [9] V. M. Shabaev et al. g factor of high-Z lithiumlike ions. Phys. Rev. A, 65: 062104, Jun 2002. doi:10.1103/PhysRevA.65.062104.
- [10] Thomas Beier. The g_j factor of a bound electron and the hyperfine structure splitting in hydrogenlike ions. *Physics Reports*, 339(2):79–213, 2000. ISSN 0370-1573. doi:https://doi.org/10.1016/S0370-1573(00)00071-5.

REFERENCES

- [11] M. Block. Recent developments in Penning-trap mass spectrometry. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 376:265–269, 2016. ISSN 0168-583X. doi:https://doi.org/10.1016/j.nimb.2016.02.027. Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11-15 May 2015.
- [12] W. Quint et al. Laser-microwave double-resonance technique for g-factor measurements in highly charged ions. *Phys. Rev. A*, 78:032517, Sep 2008. doi:10.1103/PhysRevA.78.032517.
- [13] H. Häffner et al. High-accuracy measurement of the magnetic moment anomaly of the electron bound in hydrogenlike carbon. *Phys. Rev. Lett.*, 85:5308–5311, Dec 2000. doi:10.1103/PhysRevLett.85.5308.
- [14] J. Verdú et al. Electronic g Factor of Hydrogenlike Oxygen ¹⁶O⁷⁺. Phys. Rev. Lett., 92:093002, Mar 2004. doi:10.1103/PhysRevLett.92.093002.
- [15] Florian Köhler et al. Isotope dependence of the Zeeman effect in lithium-like calcium. Nature communications, 7:10246, 01 2016. doi:10.1038/ncomms10246.
- [16] A. Wagner et al. g Factor of Lithiumlike Silicon ${}^{28}Si^{11+}$. Phys. Rev. Lett., 110: 033003, Jan 2013. doi:10.1103/PhysRevLett.110.033003.
- [17] David Lindenfels et al. Half-open Penning trap with efficient light collection for precision laser spectroscopy of highly charged ions. *Hyperfine Interactions*, 227, 02 2014. doi:10.1007/s10751-013-0961-z.
- [18] David von Lindenfels et al. Bound electron g-factor measurement by doubleresonance spectroscopy on a fine-structure transition. Canadian Journal of Physics, 89(1):79–84, 2011. doi:10.1139/P10-071.
- [19] Klaus Blaum. High-accuracy mass spectrometry with stored ions. *Physics Reports*, 425(1):1–78, 2006. ISSN 0370-1573. doi:https://doi.org/10.1016/j.physrep.2005.10.011.
- [20] S. Eliseev et al. A phase-imaging technique for cyclotron-frequency measurements. Applied Physics B: Lasers and Optics, 114:107–128, Sept 2013. doi:10.1007/s00340-013-5621-0.

- [21] S. Eliseev et al. Phase-Imaging Ion-Cyclotron-Resonance Measurements for Short-Lived Nuclides. *Phys. Rev. Lett.*, 110:082501, Feb 2013. doi:10.1103/PhysRevLett.110.082501.
- [22] R. S. Sidhu. Measurement of the bound-state beta decay of bare 205Tl81+ ions at the ESR. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2021.
- [23] Miodrag K. Pavićević et al. Lorandite from Allchar as geochemical detector for pp-solar neutrinos. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 895: 62-73, 2018. ISSN 0168-9002. doi:https://doi.org/10.1016/j.nima.2018.03.039.
- [24] M.K. Pavićević et al. New data for the geochemical determination of the solar pp-neutrino flux by means of lorandite mineral. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 621(1):278–285, 2010. ISSN 0168-9002. doi:https://doi.org/10.1016/j.nima.2010.06.090.
- [25] S. Earnshaw. On the nature of the molecular forces which regulate the constitution of the luminiferous ether. *Trans. Cambridge Philos. Soc.*, 1842.
- [26] W. Paul et al. Ein Neues Massenspektrometer Ohne Magnetfeld. Zeitschrift Naturforschung Teil A, 8:448, 07 1953. doi:10.1515/zna-1953-0710.
- [27] H. G. Dehmelt et al. "Bolometric" Technique for the rf Spectroscopy of Stored Ions. Phys. Rev. Lett., 21:127–131, Jul 1968. doi:10.1103/PhysRevLett.21.127.
- [28] F. M. Penning. Glow discharge between coaxial cylinders at low pressures in an axial magnetic field. *Physica (Utrecht)*, 3:873, 1936.
- [29] J.R. Pierce. Theory and Design of Electron Beams. D. Van Nostrand Co., Inc., 1954. ISBN 1124091300.
- [30] Randall D. Knight. The general form of the quadrupole ion trap potential. *International Journal of Mass Spectrometry and Ion Physics*, 51(1):127–131, 1983. ISSN 0020-7381. doi:https://doi.org/10.1016/0020-7381(83)85033-5.
- [31] G. Gabrielse et al. Cylindrical Penning traps with orthogonalized anharmonicity compensation. International Journal of Mass Spectrometry and Ion Processes, 57(1):1–17, 1984. ISSN 0168-1176. doi:https://doi.org/10.1016/0168-1176(84)85061-2.

- [32] G. Gabrielse et al. Open-endcap Penning traps for high precision experiments. *International Journal of Mass Spectrometry and Ion Processes*, 88(2):319–332, 1989. ISSN 0168-1176. doi:https://doi.org/10.1016/0168-1176(89)85027-X.
- [33] Joseph Tan et al. One electron in an orthogonalized cylindrical Penning trap. Applied Physics Letters, 55(20):2144–2146, 1989. doi:10.1063/1.102084.
- [34] D. von Lindenfels. Experimental Studies of Highly Charged Ions in a Penning Trap for the Measurement of Electron Magnetic Moments by Double-Resonance Spectroscopy. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2015.
- [35] M. Vogel. Particle Confinement in Penning Traps: An Introduction (Springer Series on Atomic, Optical, and Plasma Physics). Springer, 2018. ISBN 978-3-319-76263-0.
- [36] Lowell S. Brown et al. Precision spectroscopy of a charged particle in an imperfect Penning trap. *Phys. Rev. A*, 25:2423–2425, Apr 1982. doi:10.1103/PhysRevA.25.2423.
- [37] Jaroslaw Labaziewicz et al. Temperature Dependence of Electric Field Noise above Gold Surfaces. *Phys. Rev. Lett.*, 101:180602, Oct 2008. doi:10.1103/PhysRevLett.101.180602.
- [38] Lowell S. Brown et al. Geonium theory: Physics of a single electron or ion in a Penning trap. Rev. Mod. Phys., 58:233–311, Jan 1986. doi:10.1103/RevModPhys.58.233.
- [39] M. Abramowitz et al. Handbook of Mathematical Functions with Formulas, Graphs, and Mathematical Tables. Dover, ninth dover printing, tenth gpo printing edition, 1964.
- [40] P. W. Anderson et al. Hard Superconductivity: Theory of the Motion of Abrikosov Flux Lines. *Reviews of Modern Physics*, 36(1):39–42, January 1964. doi:10.1103/RevModPhys.36.39.
- [41] W. Quint and M. Vogel, editors. Fundamental Physics in Particle Traps. Springer Berlin Heidelberg, 2014. ISBN 978-3-642-45200-0.

- [42] Martin Kretzschmar. Theory of the elliptical Penning trap. International Journal of Mass Spectrometry, 275(1):21–33, 2008. ISSN 1387-3806. doi:https://doi.org/10.1016/j.ijms.2008.05.009.
- [43] M. S. Ebrahimi. Resistive Cooling of Highly- Charged-Ion Ensembles for Precision Spectroscopy in the ARTEMIS Penning Trap. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2018.
- [44] G. Bollen et al. The accuracy of heavy-ion mass measurements using time of flight-ion cyclotron resonance in a Penning trap. Journal of Applied Physics, 68(9):4355–4374, 1990. doi:10.1063/1.346185.
- [45] G. Bollen et al. Resolution of nuclear ground and isomeric states by a Penning trap mass spectrometer. *Phys. Rev. C*, 46:R2140–R2143, Dec 1992. doi:10.1103/PhysRevC.46.R2140.
- [46] J.B. Jeffries et al. Theory of space-charge shift of ion cyclotron resonance frequencies. International Journal of Mass Spectrometry and Ion Processes, 54(1):169–187, 1983. ISSN 0168-1176. doi:https://doi.org/10.1016/0168-1176(83)85016-2.
- [47] K. Blaum et al. Recent developments at ISOLTRAP: towards a relative mass accuracy of exotic nuclei below 10⁸. Journal of Physics B: Atomic, Molecular and Optical Physics, 36(5):921–930, feb 2003. doi:10.1088/0953-4075/36/5/311.
- [48] M. König et al. Quadrupole excitation of stored ion motion at the true cyclotron frequency. International Journal of Mass Spectrometry and Ion Processes, 142(1):95–116, 1995. ISSN 0168-1176. doi:https://doi.org/10.1016/0168-1176(95)04146-C.
- [49] Wayne Itano et al. Cooling Methods in Ion Traps. *Physica Scripta*, 1995:106, 01 2007. doi:10.1088/0031-8949/1995/T59/013.
- [50] D. J. Wineland et al. Principles of the stored ion calorimeter. Journal of Applied Physics, 46(2):919–930, 1975. doi:10.1063/1.321602.
- [51] D. H. E. Dubin. Trapped Charged Particles. World Scientific, 2016.
- [52] B. M. Penetrante et al. Modeling the ion-source performance of an electronbeam ion trap (invited). *Review of Scientific Instruments*, 63(4):2806–2811, 1992. doi:10.1063/1.1142812.

- [53] H.W. Ellis et al. Transport properties of gaseous ions over a wide energy range. Part III. Atomic Data and Nuclear Data Tables, 31(1):113–151, 1984. ISSN 0092-640X. doi:https://doi.org/10.1016/0092-640X(84)90018-4.
- [54] G. Savard et al. A new cooling technique for heavy ions in a Penning trap. *Physics Letters A*, 158(5):247–252, 1991. ISSN 0375-9601. doi:https://doi.org/10.1016/0375-9601(91)91008-2.
- [55] C. Weber. Konzeption eines kryogenen Penningfallenaufbaus fuer SHIPTRAP und Massenbestimmungen von Radionukliden um den Z = 82 Schalenabschluss an ISOLTRAP. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2004.
- [56] B. Andelic. Direct mass measurements of No, Lr and Rf isotopes with SHIP-TRAP and developments for chemical isobaric separation. PhD thesis, University of Valencia, 2003.
- [57] Block, M. et al. Towards direct mass measurements of nobelium at SHIPTRAP. Eur. Phys. J. D, 45(1):39–45, 2007. doi:10.1140/epjd/e2007-00189-2.
- [58] S.Ulmer. First Observation of Spin Flips with a Single Proton Stored in a Cryogenic Penning Trap. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2011.
- [59] H. Nagahama et al. Highly sensitive superconducting circuits at 700 kHz with tunable quality factors for image-current detection of single trapped antiprotons. *Review of Scientific Instruments*, 87(11):113305, 2016. doi:10.1063/1.4967493.
- [60] V. N. Gheorghe et al. Charged Particle Traps. Springer-Verlag, Heidelberg, 2004. ISBN 3540220437.
- [61] P. K. Ghosh. Ion Traps. Oxford university press, 1996. ISBN 0198539959.
- [62] American radio relay league. American radio relay league handbook for radio amateurs 1994. American radio relay league, 1993. ISBN 0-87259-171-9.
- [63] Stefan Ulmer et al. High-Precision Comparison of the Antiprotonto-Proton Charge-To-Mass Ratio. Nature, pages 196–199, 08 2015. doi:10.1038/nature14861.

- [64] Martez Ahmadi et al. Observation of the hyperfine spectrum of antihydrogen. Nature, 548:66–69, 08 2017. doi:10.1038/nature23446.
- [65] F. Heisse et al. High-Precision Measurement of the Proton's Atomic Mass. Phys. Rev. Lett., 119:033001, Jul 2017. doi:10.1103/PhysRevLett.119.033001.
- [66] Klaus Blaum et al. Precision atomic physics techniques for nuclear physics with radioactive beams. *Physica Scripta*, 2013(T152):014017, jan 2013. doi:10.1088/0031-8949/2013/T152/014017.
- [67] Julian Schwinger. On Quantum-Electrodynamics and the Magnetic Moment of the Electron. Phys. Rev., 73:416–417, Feb 1948. doi:10.1103/PhysRev.73.416.
- [68] Julian Breit. The magnetic moment of the electron. Nature, 122:649–649, 1928. doi:10.1038/122649a0.
- [69] A. V. Nefiodov et al. Nuclear-Polarization Correction to the Bound-Electron g Factor in Heavy Hydrogenlike Ions. *Phys. Rev. Lett.*, 89:081802, Aug 2002. doi:10.1103/PhysRevLett.89.081802.
- [70] A. Mooser et al. Demonstration of the double Penning Trap technique with a single proton. *Physics Letters B*, 723(1):78-81, 2013. ISSN 0370-2693. doi:https://doi.org/10.1016/j.physletb.2013.05.012.
- [71] Takashi Nakamura et al. Precision spectroscopy of the Zeeman splittings of the 9Be+22S1/2 hyperfine structure for nuclear structure studies. Optics Communications, 205(4):329–336, 2002. ISSN 0030-4018. doi:https://doi.org/10.1016/S0030-4018(02)01259-2.
- [72] Jean Brossel et al. A New "Double Resonance" Method for Investigating Atomic Energy Levels. Application to Hg³P₁. Phys. Rev., 86:308–316, May 1952. doi:10.1103/PhysRev.86.308.
- [73] H. F. Beyer. Introduction to the Physics of Highly Charged Ions. Taylor and Francis Ltd., 2002.
- [74] V Mäckel et al. Laser spectroscopy of highly charged argon at the Heidelberg electron beam ion trap. *Physica Scripta*, 2013(T156):014004, sep 2013. doi:10.1088/0031-8949/2013/T156/014004.

- [75] A. Lapierre et al. Relativistic Electron Correlation, Quantum Electrodynamics, and the Lifetime of the $1s^22s^22p\ ^2P^o_{3/2}$ Level in Boronlike Argon. *Phys. Rev. Lett.*, 95:183001, Oct 2005. doi:10.1103/PhysRevLett.95.183001.
- [76] Stefan Borneis et al. Ground-state hyperfine structure of heavy hydrogen-like ions. *Hyperfine Interactions*, 127:305–310, August 2000. doi:10.1023/A:1012676508596.
- [77] Volker Hannen et al. Lifetimes and g-factors of the HFS states in H-like and Li-like bismuth. Journal of Physics B: Atomic, Molecular and Optical Physics, 52(8):085003, apr 2019. doi:10.1088/1361-6455/ab0ef0.
- [78] C. F. v. Weizsäcker. Zur Theorie der Kernmassen. Zeitschrift fuer Physik, 96: 431–458, 1935.
- [79] O. Haxel et al. On the Magic numbers in nuclear structure. *Physical Review*, 75:1766–1766, 1949.
- [80] M. G. Mayer. On closed shells in nuclei. *Physical Review*, 74:235–239, 1935.
- [81] R. D. Woods et al. Diffuse Surface Optical Model for Nucleon-Nuclei Scattering. *Physical Review*, 95:577–578, 1954.
- [82] A. Sobiczewski et al. Description of structure and properties of superheavy nuclei. Progress in Particle and Nuclear Physics, 58:292–349, 2007.
- [83] M. Eibach et al. Direct high-precision mass measurements on 241,243 Am, 244 Pu, and 249 Cf. *Phys. Rev. C*, 89:064318, Jun 2014. doi:10.1103/PhysRevC.89.064318.
- [84] J. Dechargé et al. Bubbles and semi-bubbles as a new kind of superheavy nuclei. Nuclear Physics A, 716:55–86, 2003. ISSN 0375-9474. doi:https://doi.org/10.1016/S0375-9474(02)01398-2.
- [85] Michael Block et al. Direct mass measurements above uranium bridge the gap to the island of stability. *Nature*, 463:785–8, 02 2010. doi:10.1038/nature08774.
- [86] E Ramirez et al. Direct Mapping of Nuclear Shell Effects in the Heaviest Elements. Science (New York, N.Y.), 337:1207–10, 08 2012. doi:10.1126/science.1225636.
- [87] O. Kaleja et al. Direct high-precision mass spectrometry of superheavy elements with SHIPTRAP. *Phys. Rev. C*, 106:054325, Nov 2022. doi:10.1103/PhysRevC.106.054325.
- [88] W.J. Huang et al. The AME 2020 atomic mass evaluation (I). Evaluation of input data and adjustment procedures. *Chinese Physics C*, 45(3):030002, mar 2021. doi:10.1088/1674-1137/abddb0.
- [89] O. Kaleja. High-precision mass spectrometry of nobelium, lawrencium and rutherfordium isotopes and studies of long-lived isomers with SHIPTRAP. PhD thesis, Johannes Gutenberg-Universität Mainz, Germany, 2020.
- [90] R.-D Herzberg et al. Nuclear isomers in superheavy elements as stepping stones towards the island of stability. *Nature*, 442:896–9, 09 2006. doi:10.1038/nature05069.
- [91] P. Filianin. Measurements of low decay energies of beta-processes using Penning traps. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2019.
- [92] D. A. Nesterenko et al. Study of radial motion phase advance during motion excitations in a Penning trap and accuracy of JYFLTRAP mass spectrometer. *Eur. Phys. J. A*, 57(11):302, 2021. doi:10.1140/epja/s10050-021-00608-3.
- [93] G. Eitel et al. Position-sensitive ion detection in precision Penning trap mass spectrometry. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 606(3):475– 483, 2009. ISSN 0168-9002. doi:https://doi.org/10.1016/j.nima.2009.04.046.
- [94] UNILAC: GSI. August 2022. URL https://www.gsi.de/en/work/ beschleunigerbetrieb/beschleuniger/unilac/unilac.
- [95] K. Blasche et al. The heavy ion synchrotron SIS. France: Editions Frontieres. B. Petit-Jean-Genaz, Ch. (Ed.), 1992. URL https://cds.cern.ch/record/ 241356.
- [96] F. Herfurth et al. The HITRAP facility for slow highly charged ions. *Physica Scripta*, T166:014065, nov 2015. doi:10.1088/0031-8949/2015/t166/014065.
- [97] J. Klimes. (to be published). PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2023.

- [98] C. Vogel. Aufbau und Charakterisierung eines Kaltventils fuer Penning-Fallen Experimente. Bachelor thesis, Technische Universität Darmstadt, Germany, 2014.
- [99] SUMITOMO pulse tube cryocooler. Specification sheet. August 2022. URL https://www.shicryogenics.com/product/ rp-082b2-4k-pulse-tube-cryocooler-series/.
- [100] Stahl Electronics. User manual. August 2021. URL https://www. stahl-electronics.com/high-voltage-source.html.
- [101] Francesco Bonaccorso et al. Graphene Photonics and Optoelectronics. Nature Photonics, 4, 06 2010. doi:10.1038/nphoton.2010.186.
- [102] Graphenea: Dedicated Graphene Foundry. March 2022. URL https://www.graphenea.com/pages/article-graphenea.
- [103] M. Wiesel. Preparation and Investigation of Highly Charged Ions in a Penning Trap for the Determination of Atomic Magnetic Moments. PhD thesis, Technische Universität Darmstadt, Germany, 2017.
- [104] D.M. Pozar. Microwave Engineering. John Wiley and Sons inc, 2011. ISBN 0470631554.
- [105] F.E. Terman. Electronic and Radio Engineering (Electrical and Electronic Engineering). McGraw-Hill Inc. US, 1955. ISBN 0070635099.
- [106] D.H. Staelin. *Electromagnetics and Applications*. Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, 2011.
- [107] S.Stahl. Aufbau eines Experiments zur Bestimmung elektronischer g-Faktoren einzelner wasserstoffaehnlicher Ionen. PhD thesis, Johannes Gutenberg-Universität Mainz, Germany, 1998.
- [108] Adrian Tae-Jin Lee. A low-power-dissipation broadband cryogenic preamplifier utilizing GaAs MESFETs in parallel. *Review of Scientific Instruments*, 64(8): 2373–2378, 1993. doi:10.1063/1.1143938.
- [109] A. M. Robinson et al. Cryogenic amplifier for MHz with a high input impedance using a commercial pseudomorphic high electron mobility transistor. *Review of Scientific Instruments*, 75(10):3169–3176, 2004. doi:10.1063/1.1790586.

- [110] L. DiCarlo et al. System for measuring auto- and cross correlation of current noise at low temperatures. *Review of Scientific Instruments*, 77(7):073906, 2006. doi:10.1063/1.2221541.
- [111] J.C. Holt. The Art of Electronics. CambridgeUniversity Press, UK, 2015. ISBN 0521809266.
- [112] A. Martin. Laser Spectroscopy Invertigations of Exotic States in Noble Gases.
 PhD thesis, Technische Universität Darmstadt, Germany, 2017.
- [113] P. Baus. Characterisation of a Wide Tunable, Robust, Multi Application Diode Laser for Spectroscopy. Master's thesis, Technische Universität Darmstadt, Germany, 2014.
- [114] Cariou J. et al. Atlas du spectre d'absorption de la molecule de tellure. Laboratoire Aime-Cotton, Orsay (France), 1980.
- [115] S. Albrecht et al. A laser system for the spectroscopy of highly charged bismuth ions. Appl. Phys. B, 107(4):1069–1074, 2012. doi:10.1007/s00340-011-4732-8.
- [116] M. Kiffer. Breitbandanregung von Ionen in einer Penningfalle. Bachelor thesis, Friedrich-Schiller-University Jena, Germany, 2016.
- [117] E. O. Brigham. The Fast Fourier Transform: An Introduction to Its Theory and Application. Prentice Hall, 1973.
- [118] H.W. Gäggeler et al. Cold fusion reactions with 48Ca. Nuclear Physics A, 502:561–570, 1989. ISSN 0375-9474. doi:https://doi.org/10.1016/0375-9474(89)90689-1.
- [119] G. Munzenberg et al. The velocity filter SHIP, a separator of unslowed heavy ion fusion products. Nucl. Instrum. Meth., 161:65–82, 1979. doi:10.1016/0029-554X(79)90362-8.
- [120] O. Kaleja et al. The performance of the cryogenic buffer-gas stopping cell of SHIPTRAP. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 463:280–285, jan 2020. ISSN 0168-583X. doi:10.1016/j.nimb.2019.05.009.
- S. Hofmann et al. The discovery of the heaviest elements. *Rev. Mod. Phys.*, 72: 733–767, Jul 2000. doi:10.1103/RevModPhys.72.733.

- [122] Seperator for Heavy Ion reaction Products (SHIP) @ GSI. June 2022. URL https://www.gsi.de/work/forschung/nustarenna/nustarenna_ divisions/she_physik/experimental_setup/ship.
- [123] C. Droese et al. The cryogenic gas stopping cell of SHIPTRAP. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 338:126–138, 11 2014. doi:10.1016/j.nimb.2014.08.004.
- [124] C. Droese. The masses of nobelium and lawrencium isotopes, the mass difference between 180W and 180Hf, and a characterization of the future cryogenic stopping cell of the online masspectrometer SHIPTRAP. PhD thesis, Ernst-Moritz-Ardnt-Universität Griefswald, Germany, 2014.
- [125] D. Rodriguez. An RFQ buncher for accumulation and cooling of heavy radionuclides at SHIPTRAP and high precision mass measurements on unstable Kr isotopes at ISOLTRAP. PhD thesis, University of Valencia, 2003.
- [126] D. A. Nesterenko et al. Direct determination of the atomic mass difference of ¹⁸⁷Re and ¹⁸⁷Os for neutrino physics and cosmochronology. *Phys. Rev. C*, 90: 042501, Oct 2014. doi:10.1103/PhysRevC.90.042501.
- [127] S. Eliseev et al. Direct Measurement of the Mass Difference of ¹⁶³Ho and ¹⁶³Dy Solves the Q-Value Puzzle for the Neutrino Mass Determination. *Phys. Rev. Lett.*, 115:062501, Aug 2015. doi:10.1103/PhysRevLett.115.062501.
- [128] S. Eliseev et al. Octupolar-Excitation Penning-Trap Mass Spectrometry for Q-Value Measurement of Double-Electron Capture in ¹⁶⁴Er. Phys. Rev. Lett., 107:152501, Oct 2011. doi:10.1103/PhysRevLett.107.152501.
- [129] S. Rahaman. First Tests of the Penning traps of the SHIPTRAP Facility. Diploma thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2002.
- [130] G. Sikler. Massenspektrometrie kurzlebiger Sr- und Sn- Isotope und Aufbau der SHIPTRAP-Penningfallen. Diploma thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2003.
- [131] S. Rahaman. First on-line mass measurements at SHIPTRAP and mass determinations of neutron-rich Fr and Ra isotopes at ISOLTRAP. PhD thesis, Ruprecht-Karls-Universität Heidelberg, Germany, 2005.

- [132] Brookhaven National Laboratory National Nuclear Data Center (NNDC). NuDat (Nuclear Structure and Decay Data). March 2008. URL https: //www.nndc.bnl.gov/nudat3/.
- [133] Jörg Runke et al. Preparation of actinide targets for the synthesis of the heaviest elements. Journal of Radioanalytical and Nuclear Chemistry, 299, 02 2014. doi:10.1007/s10967-013-2616-6.
- [134] N. Trautmann et al. Preparation of actinide targets by electrodeposition. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 282(1):102–106, 1989. ISSN 0168-9002. doi:https://doi.org/10.1016/0168-9002(89)90117-4.
- [135] Alessio Vascon et al. The performance of thin layers produced by molecular plating as α-particle sources. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 721:35–44, 09 2013. doi:10.1016/j.nima.2013.04.050.
- [136] P. Schwamb. Entwicklung eines Detektors zum direkten massenselektiven Ionennachweis nach Resonanzionisationsspektroskopie in einer Puffergaszelle. PhD thesis, Johannes Gutenberg-Universität Mainz, Germany, 1996.
- [137] ORTEC ULTRA and ULTRA AS. User Manual, Ion-Implanted-Silicon Charged-Particle Detectors, May 2022. URL https://www.ortec-online. com/-/media/ametekortec/brochures/u/ultra.pdf?la=en&revision= 88b06c5e-cacd-480b-bfdb-aa358ffd6fd4.
- [138] P. Filianin et al. The decay energy of the pure s-process nuclide 123Te. *Physics Letters B*, 758:407–411, 2016. ISSN 0370-2693. doi:https://doi.org/10.1016/j.physletb.2016.04.059.
- [139] Olha Bodnar et al. On the adjustment of inconsistent data using the Birge ratio. Metrologia, 51(5):516, aug 2014. doi:10.1088/0026-1394/51/5/516.
- [140] Chaudhuri, A. et al. Carbon-cluster mass calibration at SHIPTRAP. Eur. Phys. J. D, 45(1):47–53, 2007. doi:10.1140/epjd/e2007-00001-5.
- [141] Kellerbauer, A. et al. From direct to absolute mass measurements: A study of the accuracy of ISOLTRAP. Eur. Phys. J. D, 22(1):53–64, 2003. doi:10.1140/epjd/e2002-00222-0.

- [142] G. Gabrielse. The true cyclotron frequency for particles and ions in a penning trap. International Journal of Mass Spectrometry, 279(2):107–112, 2009. ISSN 1387-3806. doi:https://doi.org/10.1016/j.ijms.2008.10.015.
- [143] M. J. Gutierrez. Private communication, 2022.
- [144] C. Droese et al. Investigation of the magnetic field fluctuation and implementation of a temperature and pressure stabilization at SHIPTRAP. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 632(1):157–163, 2011. ISSN 0168-9002. doi:https://doi.org/10.1016/j.nima.2010.12.176.
- [145] R. J. Walen. Spectrographic α Due Radium 224 et de Ses Derives. *Compt.Rend.*, 255:1604, 1962.
- [146] G. Bastin-Scoffier. Niveaux du Francium 221 Observes a Partir de l'Actinium 225. Compt.Rend., 265B:863, 1967.
- [147] B. S. Dzhelepov et al. Alpha Decay of ²²⁵Ac, ²²¹Fr and ²¹⁷At. *Izv.Akad.Nauk*, SSSR:Ser.Fiz. 31, 568, 1967.
- [148] C. F. Leang et al. Etude du schema de niveaux de l'astatine 217. Compt.Rend., 266B:629, 1968.
- [149] O. Sackur. Ueber die Zerfallsconstante der Radiumemanation. Ber.Dtsch. Chem. Ges., 38:1753, 1905. doi:10.1002/cber.19050380287.
- [150] R. J. Walen et al. Spectrographie α de ²²³Ra (AcX) et Ses Derives. Nuclear Physics, 35:232, 1962. ISSN 0029-5582. doi:10.1016/0029-5582(62)90109-8.
- [151] B. Grennberg et al. Absolute Measurements of α -Ray Energies. *Metrologia*, 7: 65, apr 1971. doi:10.1088/0026-1394/7/2/005.
- [152] A. A. Vorobev et al. Measurement of the Energy of Alpha Particles from Some Emitters. *Zhur.Eksptl.i Teoret.Fiz.*, 39:70, 1960. URL https://www.osti.gov/ biblio/4145906.
- [153] K. Valli. An Experimental Investigation of the Alpha Fine Structure in Ac-225, Fr-221, At-217, and Po-213. Ann. Acad. Sci. Fennicae, pages Ser.A VI, No.165, 1 1964. URL https://www.osti.gov/biblio/4661412.

- [154] T. Vylov et al. The Decay of ²²¹Rn. Izv.Akad.Nauk, SSSR:Ser.Fiz. 41, 1635, 1977.
- [155] J. David Bowman et al. Alpha spectroscopy of nuclides produced in the interaction of 5 GeV protons with heavy element targets. *Phys. Rev. C*, 25:941–951, Feb 1982. doi:10.1103/PhysRevC.25.941.
- [156] H. Van Krugten et al. Beta continuum measurements on 226 ac and daughter products of 225 ac. *Physica*, 40:253, 1968. ISSN 0031-8914. doi:https://doi.org/10.1016/0031-8914(68)90022-0.
- [157] C. R. Cothern et al. Studies in the Active Deposit of Actinium. Part II: The Decay of ²¹¹Pb (AcB). Can.J.Phys., 43:383, 1965. URL https://doi.org/10. 1139/p65-035.
- [158] A. Sperduto et al. Q-Value Measurements at M.I.T. In Walter H. Johnson, editor, *Proc.Intern.Conf.Nuclidic Masses*, pages 289–304, Vienna, 1964. Springer Vienna. ISBN 978-3-7091-5556-1.
- [159] G. Muehllehner et al. ²⁰⁸Pb(d, p) and (d, t) Reactions with 15- to 25-MeV Deuterons. *Phys.Rev.*, 159:1039, Jul 1967. doi:10.1103/PhysRev.159.1039.
- [160] H. Behrens et al. The First Forbidden Beta Decay of Pb²⁰⁹. Zeitschrift für Physik A Hadrons and nuclei, 252:349–361, 1972. URL https://doi.org/10. 1007/BF01387305.
- [161] J. J. Kolata et al. Shell-Model Structure of ²¹⁰Bi: ²⁰⁹Bi(d, p) at 17 MeV. Phys. Rev. C, 5:568–578, 1972. doi:10.1103/PhysRevC.5.568.
- [162] D.G. Kovar et al. Excitation of two-particle-one-hole states by the 208Pb(d, p)209Pb reaction. Nuclear Physics A, 231(2):266–300, 1974. ISSN 0375-9474. doi:https://doi.org/10.1016/0375-9474(74)90522-3.
- [163] A. Såmark-Roth et al. Low-lying states in ²¹⁹Ra and ²¹⁵Rn: Sampling microsecond α -decaying nuclei. *Phys. Rev. C*, 98:044307, Oct 2018. doi:10.1103/PhysRevC.98.044307.
- [164] A. Rytz. Absolute Measurement of the Energy of the Most Important Natural Alpha Emitters. *Helvetica Physica Acta (Switzerland)*, 34:240, 1 1961. URL https://www.osti.gov/biblio/4030176.

- [165] W. F. Davidson et al. Studies in the Decay of the Active Deposit of Actinium.
 III. Levels in ²¹¹Bi and its Daughter Products. *Can.J.Phys.*, 45:2295, 1967.
 doi:https://doi.org/10.1139/p67-183.
- [166] Martin Kretzschmar. Theory of simultaneous dipole and quadrupole excitation of the ion motion in a Penning trap. International Journal of Mass Spectrometry, 325-327:30–44, 2012. ISSN 1387-3806. doi:https://doi.org/10.1016/j.ijms.2012.07.015. Eugene N. Nikolaev 65th Birthday Honor Issue.
- [167] Martin Kretzschmar. The Ramsey method in high-precision mass spectrometry with Penning traps: Theoretical foundations. International Journal of Mass Spectrometry, 264(2):122–145, 2007. ISSN 1387-3806. doi:https://doi.org/10.1016/j.ijms.2007.04.002.
- [168] Martin Kretzschmar. On the phase dependence of the interconversion of the motional modes in a Penning trap by quadrupolar excitation. International Journal of Mass Spectrometry, 309:30–38, 2012. ISSN 1387-3806. doi:https://doi.org/10.1016/j.ijms.2011.08.022.
- [169] Vibrational states of 207Tl. Nuclear Physics A, 127(1):71-80, 1969. ISSN 0375-9474. doi:https://doi.org/10.1016/0375-9474(69)90768-4.
- [170] T. J. Kennett et al. The Level Structure of ¹⁴²Pr Deduced from the ¹⁴¹Pr(n, γ)¹⁴²Pr Reaction. Can. J. Phys., 59:1212, 1981. doi:https://doi.org/10.1139/p81-159.
- [171] L.A. Sushkov et al. Leningradskiy Institut Yadernoi Fiziki 644, 1981.
- [172] P. Hungerford et al. Neutron Binding and Excitation Energies of some Pb Isotopes. Zeitschrift für Physik A Hadrons and nuclei, A313:349, 1983. doi:https://doi.org/10.1007/BF01439488.
- [173] T. Belgya et al. Levels of ²⁰⁸Pb from the ²⁰⁷Pb (n, γ) reaction with a guided neutron beam. *Phys. Rev. C*, 57:2740–2743, May 1998. doi:10.1103/PhysRevC.57.2740.
- [174] R.B. Firestone et al. Thermal neutron capture cross sections and neutron separation energies. Database of prompt gamma rays from slow neutron capture for elemental analysis - Vienna : International Atomic Energy Agency. IAEA-Library 45, 2006. URL https://www-nds.iaea.org/pgaa/tecdoc.pdf.

- [175] Kathrin Kromer et al. High-precision mass measurement of doubly magic ²⁰⁸Pb.
 Eur. Phys. J. A, 58(10):202, 2022. doi:10.1140/epja/s10050-022-00860-1.
- [176] Alan G. Marshall et al. Fourier transform ion cyclotron resonance mass spectrometry: A primer. Mass Spectrometry Reviews, 17(1):1– 35, 1998. doi:https://doi.org/10.1002/(SICI)1098-2787(1998)17:1<1::AID-MAS1>3.0.CO;2-K.
- [177] M. Shaaban. Preparing and testing the detection system for the ARTEMIS experiment. Master's thesis, Technische Universität Darmstadt, Germany, 2018.

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