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Direct mass measurements of No, Lr and Rf isotopes with SHIPTRAP and developments for chemical isobaric separation

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Direct mass measurements of No, Lr and Rf
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chemical isobaric separation

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Cover: cartoon showing mass measurements of the heaviest elements

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Direct mass measurements of No, Lr and Rf isotopes with SHIPTRAP and developments for chemical isobaric separation

PhD thesis

to obtain the degree of PhD at the
University of Groningen
on the authority of the
Rector Magnificus Prof. C. Wijmenga
and in accordance with
the decision by the College of Deans.

This thesis will be defended in public on

Friday 9 July 2021 at 12.45 hours

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To my parents, Gospova and Slavko

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Introduction

Every nucleus is characterized by its numbers of protons Z and neutrons N which give unique nuclear properties to it. Depending on these numbers, the atomic nucleus can be stable or it decays (see Fig. 1.1) in order to obtain an energetically preferred configuration. In the focus of contemporary theoretical and experimental studies in the field of nuclear structure and nuclear astrophysics are the so-called *exotic nuclei*. They are short-lived, do not exist naturally on earth, and must be produced at reactor- and accelerator-based facilities. Experimental studies of exotic nuclei provide knowledge about their properties, which is crucial to answer questions concerning the origin and composition of matter.

The mass of the atomic nucleus is one of its basic properties. It summarizes all interactions of protons and neutrons within a nucleus. From the experimentally obtained mass $m(^A\text{X})$ of a nucleus X with the mass number $A = N + Z$, the nuclear binding energy $B(Z, N)$ can be determined. It is calculated as the difference between its mass and the sum of the masses of its constituents

Figure 1.1: The nuclear chart containing the known nuclei, arranged according to their neutron and proton numbers. Black squares correspond to the stable nuclei which are surrounded by the radioactive ones. The black vertical and horizontal lines indicate the locations of the nuclei with increased stability, the so-called magic nuclei. The encircled regions correspond to the nuclei which are of relevance for this work. Figure adapted from Ref. [1].

$$B(Z, N) = [Zm_p + Nm_n - m(^A\text{X})]c^2, \quad (1.1)$$

where m_p and m_n represent the masses of proton and neutron, respectively and c is the speed of light. The nuclear binding energy is a measure of how much energy is needed to separate the nucleus into its single constituents and, therefore, it represents a unique measure of the stability of a nucleus. Detailed studies of nuclear binding energies are important to reveal enhanced stability and the boundaries of nuclear existence. In nuclear astrophysics, differences between the masses of two nearby nuclei are used in order to understand the synthesis of different chemical elements through nucleon-capture processes.

Over the last century, several techniques have been developed for direct mass measurements. To extract the nuclear binding energy and to probe nuclear structure and astrophysical processes, a mass precision on the order of $\delta m/m = 10^{-6} - 10^{-8}$ and $\delta m/m = 10^{-7}$, respectively, is required [2]. Such precision is routinely achieved in Penning trap mass spectrometry. Penning traps use a homogeneous magnetic field to confine charged particles radially, while the axial confinement is achieved by a quadrupolar electrostatic field. A mass measurement in a Penning trap is based on the measurement of the cyclotron frequency that is an intrinsic and specific characteristic of a trapped charged particle and gives access to its mass-over-charge ratio. Different measurement techniques have been implemented at Penning-trap facilities for cyclotron-frequency measurements. Recent developments of the Phase-Imaging Ion-Cyclotron-Resonance (PI-ICR) technique at the Penning trap mass spectrometer SHIPTRAP located at the GSI accelerator facility in Darmstadt, Germany, opened the door to very accurate mass measurements at the $\delta m/m = 10^{-9}$ level even for short-lived nuclides with half-lives well below a second [3, 4]. These developments provide important contributions toward studies of nuclei far from stability.

In this work, nuclei at the extremes of nuclear stability are addressed at the upper limit of the nuclear chart indicated in Fig. 1.1 through Penning trap mass spectrometry, which has already paved the way to a better understanding of nuclear structure in these exotic species [5–7]. Within the FAIR Phase-0 program in 2018, high-precision mass spectrometry of $^{251,254}\text{No}$ ($Z = 102$) and $^{254-256}\text{Lr}$ ($Z = 103$) was performed at SHIPTRAP. The masses of ^{251}No and ^{254}Lr have been directly measured for the first time with a precision on the order of 10^{-8} . Additionally, the first direct mass measurement of a superheavy element, ^{257}Rf ($Z = 104$), performed within this work proved the feasibility of the PI-ICR technique in the region of the heaviest elements with the lowest yields (1 event detected every 10 hours) and opened the door to the direct mass measurements of superheavy elements.

In parallel to the efforts focused on studies of these elements, technical developments with the aim to access neutron-deficient isotopes in the

region of ^{100}Sn are ongoing (see Fig. 1.1). This region is of particular interest to study the nuclear structure and in particular nucleon interactions in the heaviest systems containing the same number of neutrons and protons $N = Z$ [8]. Besides, high-precision mass measurements are needed to address important astrophysical processes [8]. Aside from low yields, the biggest challenge in these studies is the separation of isobaric contaminants. These represent different chemical elements that have the same mass number as the nuclide of interest. This work presents a novel chemical separation technique and technical developments directed towards its implementation at SHIPTRAP.

This thesis is divided into 9 chapters. Chapter 2 focuses on the main physical processes that concern the production of exotic nuclei in fusion-evaporation reactions, their separation and preparation for low-energy experiments such as Penning trap mass spectrometry, which is described within the chapter. In Chapter 3, an overview of the SHIPTRAP facility is given. Chapter 4 describes the motivation for high-precision mass measurements in the region of the heaviest elements and gives an overview of the SHIPTRAP experimental campaign performed in 2018. The data that were collected during this experimental campaign have been analyzed according to the procedure presented in Chapter 5. The results are presented and discussed in Chapter 6. Chapter 7 describes how mass spectrometry in the region of ^{100}Sn can contribute to nuclear structure and nuclear astrophysics studies. The main challenges concerning isobaric separation are introduced and a novel chemical separation technique is proposed as a potential solution. To develop this technique, a new gas catcher has been built and is currently used for off-line investigations of gas-phase chemistry before being implemented at SHIPTRAP. The design of the gas catcher and the status are given in Chapter 8, followed by the summary and outlook in Chapter 9.

From isotope production to high-precision mass measurements

The choice of the most suitable nuclear reaction to produce exotic nuclei depends on the region of interest in the nuclear chart and the available technical possibilities. For instance, to produce neutron-deficient exotic nuclei up to the proton drip-line, the fusion-evaporation reactions are well suited. Moreover, this is presently the only viable option to produce superheavy nuclides.

In complete fusion-evaporation reactions, different nuclides are produced according to the most probable exit channels for the de-excitation of the compound nucleus. In order to study a specific reaction product, the separation from the primary beam is crucial and can be achieved by employing electromagnetic recoil separators. The separated fusion-evaporation reaction products have recoil energies on the order of tens of MeV. To study these nuclei through low-energy experiments, such as Penning-trap mass spectrometry, which is in the focus of this thesis, it is required to reduce their kinetic energy down to a few eV. For

this reason, gas-catcher devices have been developed to thermalize and extract the reaction products as ions.

This chapter describes the isotope production via fusion-evaporation reactions and the fundamental physics processes that occur during thermalization and extraction of ions from a gas catcher. Since the results presented in the thesis are obtained using a Penning trap mass spectrometer, the ion motion and manipulation in a Penning trap is discussed as well as the technique that is used for the mass determination.

2.1 Production of exotic nuclei in fusion-evaporation reactions

In a fusion-evaporation reaction an accelerated projectile bombards a stationary target with an energy that is sufficient to overcome the Coulomb repulsion between the two nuclei. This may lead to the formation of a di-nuclear system in the capture stage. At this point, the individual nucleons collide with each other and thermalize, which can result either in quasi-fission or fusion into a compound nucleus [9–11]. In the case of quasi-fission, a projectile-like fragment and a target-like

Figure 2.1: Schematic of the fusion-evaporation reaction. For further details, see text.

fragment are formed; see Fig. 2.1. When instead the two colliding nuclei fuse, their compound system is in a highly excited state. It de-excites either via fission or formation of evaporation residue. The latter occurs when, due to statistical fluctuations, one or more nucleons acquire sufficient energy to escape, and the compound nucleus either emits particles, typically protons, neutrons or alpha particles, or it radiates gamma rays. As the process is similar to the evaporation of molecules from a hot liquid, this stage is called evaporation. The time for the system to reach statistical equilibrium depends on the nuclear species, the type of projectile, and its energy, but it is typically on the order of 10^{-14} s. The properties of the compound nucleus determine its subsequent behavior, which means that its decay probability depends only on the total energy inside the system. In other words, the compound nucleus “forgets” the process of formation and its decay is governed solely by statistical rules [9, 10].

The cross section of the reaction $P+T \rightarrow \text{CN}^* \rightarrow A+B$, where P is the projectile, T is the target nucleus and A and B are the heavier and lighter reaction residues, respectively, is a function of the excitation energy E_{exc} and can be written as

$$\sigma_{AB}(E_{\text{exc}}) = \sigma(\text{CN}^*)(E_{\text{exc}}) \cdot P_A(E_{\text{exc}}), \quad (2.1)$$

where $\sigma(\text{CN}^*)$ is the cross section for the formation of the compound nucleus CN^* , while $P_A(E_{\text{exc}})$ is the probability to decay into the channel in which particle A is emitted. The excitation energy of the compound nucleus E_{exc} represents the difference in the total energy before the collision and the ground state energy of the created compound nucleus CN [12]

$$E_{\text{exc}} = m_P c^2 + m_T c^2 + E_P - m_{\text{CN}} c^2, \quad (2.2)$$

where m_P , m_T and m_{CN} are the masses of the projectile, the target and the compound nucleus, respectively, c is the speed of light and E_P is

the projectile kinetic energy (beam energy). Therefore, the cross section for a particular reaction can be maximized by proper selection of the projectile energy. As an example, the cross sections for production of the lawrencium isotopes $^{254-256}\text{Lr}$ calculated by the statistical model code HIVAP [13] are shown in Fig. 2.2 as a function of excitation energy. The most probable decay channel of the compound nucleus $^{257}\text{Lr}^*$ produced via the fusion-evaporation reaction of ^{48}Ca and ^{209}Bi is determined by its excitation energy. For each reaction, the cross section increases to a maximum and then decreases since at higher excitation energies it is more likely for an additional neutron to be emitted.

Figure 2.2: Excitation functions for the fusion-evaporation reactions $^{209}\text{Bi}(^{48}\text{Ca}, xn)$ in which the lawrencium isotopes $^{254-256}\text{Lr}$ are produced. The excitation functions are calculated using the HIVAP code [13].

The kinetic energy of a fusion-evaporation product E_A can be derived from the energy and momentum conservation

$$E_A = \left(\frac{m_P}{m_P + m_T} \right)^2 \cdot \frac{E_P}{m_P} \cdot m_A, \quad (2.3)$$

where m_A is the mass of the fusion-evaporation product A. E_A is on

the order of a few 100 keV/u, which is significantly lower than the projectile energy. The reaction products that recoil out of a thin target are emitted in a forward direction and due to their difference in velocity with respect to the primary beam, they are suited for kinematic separation by a velocity filter (see Section 3.1). The separation is performed by electric and magnetic fields and it is independent of the mass-over-charge ratio. To enable efficient focusing it is necessary to narrow down the broad charge-state distribution of the reaction residues. For this reason a thin carbon charge-exchange foil is usually used downstream from the target. The average charge state of heavy ions follows the distribution [14]

$$\bar{q} = Z \left[1 + \left(\frac{v}{Z^{\alpha} v'} \right)^{-1/k} \right]^{-k}, \quad (2.4)$$

where $k = 0.6$, $\alpha = 0.45$, and $v' = 3.6 \cdot 10^6$ m/s. It depends on the atomic number Z of the fusion-evaporation reaction product as well as on its velocity v . The shell structure of reaction products introduces deviations of the experimental values of $\bar{q}$ from the value given by Eq. 2.4. For ions with $Z \geq 20$ these deviations of $\bar{q}$ do not exceed 5% [14].

2.2 Ion preparation for high-precision mass measurements

To enable high-precision mass measurements, the fusion-evaporation products have to be slowed down through interactions with solid matter and gases, as for instance the thin entrance window foil and the buffer gas of a gas-catcher device (for technical details, see Section 3.2 and Chapter 8). Electric fields and a gas-flow can be used to extract and transfer ions towards a mass spectrometer. In this section the energy loss and the motion of the ions inside the gas catcher are described together with the gas-phase chemical processes.

2.2.1 Nuclear and electronic stopping

The stopping power of a medium S is defined as the average energy loss per unit path length of the ion

$$S = \frac{dE}{dx}, \quad (2.5)$$

where E stands for the kinetic energy of the ion and x is a distance along the ion trajectory in the medium.

There are three contributions to the energy loss: radiative energy loss, nuclear energy loss and electronic energy loss. The radiative energy loss is due to the emission of bremsstrahlung and it can be neglected in the case of non-relativistic ions. The nuclear energy loss appears due to elastic collisions of the ions with the nuclei in the medium. The electronic energy loss happens due to the excitation or ionization of the atoms in the medium or the projectile. Therefore, for non-relativistic ions, the total stopping power $S_t(E)$ is the sum of the nuclear stopping power $S_n(E)$ and the electronic stopping power $S_e(E)$

$$S_t(E) = S_e(E) + S_n(E). \quad (2.6)$$

The electronic energy loss is dominant at energies in the region of MeV. Therefore, this type of energy loss occurs mostly in the interactions of the ions with the atoms of the entrance window. The nuclear energy loss becomes dominant once the ions slow down to energies of several 10 keV/u and below.

Electronic energy loss

The electronic energy loss is dependent on the electronic structure of the ions and the atoms of the stopping medium. To describe the electronic stopping, different analytical approximations are introduced for the low- and the high-energy regions.

In the case when the velocity of the ion is large compared to the Bohr velocity of the electrons of the atoms in the stopping medium, the Bethe-Bloch formula [15, 16] can be applied

$$S_e = 4\pi \frac{Z_1^2 Z_2^2 e^4}{m_e v_1^2} \ln \left(\frac{2m_e v_1^2}{I} \right), \quad (2.7)$$

with Z_1 and Z_2 being the atomic numbers of the ion and the stopping medium, respectively, m_e being the mass of the electron, v_1 being the velocity of the ion, e the elementary charge and I the mean ionization potential. This process is treated as Coulomb scattering between the ion and the electrons of the stopping medium. Equation 2.7 is only valid for energies at which relativistic effects do not contribute.

At low energies of the projectile ion (keV region), the outer electrons contribute more to the electronic energy loss. An approximation, that is based on the interaction of the projectile ion with the free electron gas, is developed by Lindhard and Scharff [17]

$$S_e = 4\pi \frac{Z_1^2 Z_2 e^4}{m_e v_1^2} c_k \left(\frac{2m_e v_1^2}{I} \right)^{\frac{3}{2}}, \quad (2.8)$$

c_k is a parameter that depends on the target material. The projectile ion will lose its energy to the point where the maximum energy that it can transfer is lower than the lowest excitation energy level of the target atom.

However, the highest electronic stopping power occurs at intermediate energies. To describe the energy loss in this region, an interpolation between the low- and the high-energy stopping power is required [18].

Nuclear energy loss

Nuclear stopping is a consequence of elastic collisions between the ion and the atoms in the stopping medium. The nuclear stopping power

$S_n(E)$ can be calculated if the repulsive potential $V(r)$ between the ion and the target atom is known. This potential corresponds to the repulsive Coulomb interaction between the nuclei. The screening of the charges by electrons has to be taken into account as well. For this reason, the repulsive Coulomb potential has to be multiplied by the screening potential Φ

$$V(r) = \frac{Z_1 Z_2 e^2}{4\pi\epsilon_0 r} \Phi\left(\frac{r}{a}\right), \quad (2.9)$$

where Z_1 and Z_2 are the atomic numbers that correspond to the ion and the target atom, e is the elementary charge, ϵ_0 is the vacuum permittivity, r represents the distance between the ion and the target nucleus and a is the screening length. There are various parametrizations of the screening potential [18–21]. One of the most commonly used $V(r)$ was proposed by Ziegler, Biersack and Littmark [18] and is called *ZBL repulsive potential*. By applying this potential, the scattering angle θ in the center of mass system is given by [18]

$$\theta = \pi - 2 \int_{r_{min}}^{\infty} \frac{b}{r^2 \sqrt{1 - \frac{V(r)}{E_c} - \frac{b^2}{r^2}}} dr, \quad (2.10)$$

where b represents the impact parameter, E_c is the relative energy, and r_{min} is the distance of minimum approach between the ion and the target nucleus. The nuclear stopping power is the average energy per unit path length transferred from the ion to the target atom when summed over all impact parameters. By taking into account energy and momentum conservation, the following expression for the nuclear stopping power can be derived [18]

$$S_n(E) = \frac{8\pi m_1 m_2 E}{(m_1 + m_2)^2} \int_0^{b_{max}} \sin^2\left(\frac{\theta}{2}\right) b db, \quad (2.11)$$

where m_1 and m_2 are the masses of the ion and the target atom, respectively.

2.2.2 Ion motion through a buffer gas in the presence of electric fields

Once the ions are stopped and thermalized inside the gas catcher, their motion is determined by the influence of the applied electric field, further collisions with the buffer gas atoms and diffusion. If an ion with charge q moves in an electrostatic field $\vec{E}$, the resulting force $\vec{F}$ will be

$$\vec{F} = q\vec{E}. \quad (2.12)$$

While moving through a buffer gas, the ions experience a damping force. If the ions are thermalized and their masses are much higher than the mass of the atoms of the buffer gas, the viscous-drag model can be applied to describe the ion motion [22]. The ion, which moves slowly through the buffer gas, interacts with many atoms at the same time polarizing the atoms. The damping force $\vec{F}_{\text{damp}}$ is given as

$$\vec{F}_{\text{damp}} = -\delta m \vec{v}, \quad (2.13)$$

where m is the mass of the ion, $\vec{v}$ is its velocity and δ is

$$\delta = \frac{q}{m} \frac{1}{K}, \quad (2.14)$$

with K being the ion mobility. The ion mobility at temperature T and the gas pressure p is defined as

$$K = K_0 \frac{T}{T_0} \frac{p_0}{p}. \quad (2.15)$$

p_0 is the standard pressure 1013 mbar and T_0 is the standard temperature 273.15 K. K_0 is the reduced ion mobility and for most elements it is well known and can be found in literature.

Also, the ions can disperse through the gas by diffusion. The driving force $\vec{f}$ is a pressure-gradient force, or in case of isothermal conditions

which apply for the gas catcher, it is given by the ion density gradient ∇n

$$\vec{f} = -\nabla p = -k_B T \nabla n, \quad (2.16)$$

where k_B is Boltzmann constant. Considering the above mentioned processes, one can write the fluid equation of motion as

$$mn \frac{d\vec{v}}{dt} = qn\vec{E} - k_B T \nabla n - \frac{q}{K} n \vec{v}, \quad (2.17)$$

where n represents the ion density. If the steady state is considered, the left side of Eq. 2.17 can be set to zero and solved with respect to $\vec{v}$

$$0 = qn\vec{E} - k_B T \nabla n - \frac{q}{K} n \vec{v}, \quad (2.18)$$

$$\vec{v} = K\vec{E} - K \frac{k_B T}{q} \frac{\nabla n}{n}, \quad (2.19)$$

$$\vec{v} = K\vec{E} - D \frac{\nabla n}{n}. \quad (2.20)$$

In Eq. 2.20 the relation between the ion mobility K and the diffusion coefficient D , known as the Nernst-Einstein relation, is used

$$K = \frac{qD}{k_B T}. \quad (2.21)$$

The total flux $\vec{j}$ of ions through the gas catcher is given by

$$\vec{j} = nK\vec{E} - D\nabla n. \quad (2.22)$$

The first term in Eq. 2.22 describes the dependence of the ion flux on the ion mobility and the electric field, while the second term represents the ion motion along the density gradient, known as Fick's law of diffusion.

Systematic studies of the extraction time for $^{219}\text{Rn}^+$ ions from a gas catcher are reported in Ref. [23]. Typical values for the ion extraction time from a gas-catcher device operated at a pressure of ~ 50 mbar at room temperature are on the order of few tens of ms. Assuming the radius of the gas catcher r_0 to be 14 cm, it is possible to calculate the typical diffusion time τ_D for ^{219}Rn ions using the following equation [24]

$$\tau_D = \frac{1}{D} \left(\frac{r_0}{2.405} \right)^2. \quad (2.23)$$

For the conditions where $p = 50$ mbar at room temperature, using Eq. 2.21 and $K_0 = 17.37$ V/cm for ^{219}Rn [25] one can calculate the diffusion coefficient and from that the diffusion time. In this example, the diffusion time is calculated to be 3.4 s, which is much longer compared to the total extraction time. Therefore, as long as a potential gradient of at least $0.5 - 1$ V/cm is applied along the gas-catcher body (see Fig. 2.3), diffusion plays a minor role during ion transport through the gas catcher.

The ions can be extracted from the gas catcher by using a gas-flow [26] or a combination of gas-flow and electric fields [27–29]. In this work, the latter method is employed to extract ions by using a RF (Radio-Frequency) funnel/carpet [27]. Both systems consist of concentric ring electrodes with a decreasing diameter towards a nozzle or exit hole. The main difference is that the funnel is cone-shaped, while in the case of the carpet the electrodes are printed on a PCB (Printed Circuit Board). More details on the technical design of the RF funnel and the RF carpet can be found in Sections 3.2 and 8.7, respectively. Nevertheless, the principle for ion extraction is the same. In addition to a DC (Direct Current) voltage gradient, a RF voltage that alternates by 180° between adjacent electrodes is applied. The purpose of the DC voltage gradient

Figure 2.3: Schematic of a gas-catcher device with arrows indicating the direction of the forces and the blue dots representing the ions. A DC gradient is applied along the body to guide the ions towards a RF carpet. The RF field of the carpet repels the ions from its surface, while the DC gradient along the carpet radially focuses the ions towards the exit hole where the gas flow takes over and sweeps the ions out.

is to drag the ions towards the exit hole or the nozzle (see Fig. 2.3). The RF voltage is applied to avoid ion losses through collisions with the ring electrodes. Assuming that a quadrupolar RF potential is created by a pair of adjacent electrodes in addition to a pair of virtual electrodes, the resulting effective field E_{eff} is derived as [30]

$$E_{\text{eff}} = \frac{qV_{\text{RF}}^2\tau_v^2}{mr_0^3(1 + \Omega^2\tau_v^2)}, \quad (2.24)$$

with V_{RF} being the RF amplitude, r_0 the half-distance between the centers of adjacent electrodes, Ω the angular frequency of the RF field and τ_v the velocity relaxation time. Considering that the pressure inside the gas catcher is several tens of mbar, the velocity relaxation time, which is the average time between collisions, is much shorter ($10^{-8} - 10^{-7}$ s) than the RF period (typically 10^{-6} s) resulting in $\Omega^2\tau_v^2 \ll 1$ and thus, the effective field can be modified to (by using Eq. 2.14)

$$E_{\text{eff}} = \frac{mK^2}{q} \frac{V_{\text{RF}}^2}{r_0^3}. \quad (2.25)$$

From the formula for the resulting repulsive force F_{RF}

$$F_{\text{RF}} = qE_{\text{eff}} = \frac{mK^2V_{\text{RF}}^2}{r_0^3}, \quad (2.26)$$

it can be concluded that the extraction efficiency depends on the ion mass and the ion mobility, as well as on the amplitude of the RF field. The requirement regarding the RF frequency is that polarity changes before an ion can hit a ring electrode. A strong dependence on the gap between electrodes favors RF carpets which can be manufactured much easier than funnels with small electrode spacing. However, a RF carpet might be a source of impurities, due to outgassing of the PCB, which can cause undesired chemical reactions inside the gas catcher. Additionally, due to a large number of electrodes arranged close together, both systems are sensitive to discharges which might occur depending on the pressure and the applied voltage. The break-down voltage as a function of the pressure and the electrode gap is given by the Paschen law [31] which has to be considered in order to avoid gas ionization and the creation of impurities.

2.2.3 Gas-phase chemistry

The highly-charged reaction products enter the volume filled with buffer gas and while they are slowing down, their charge state reduces in the collisions with the atoms of buffer gas B due to charge-exchange processes such as

where R is the ion of interest and q is the charge state.

The cross section to capture an electron decreases with increasing ionization potential of the buffer gas. This is one of the reasons why helium

and argon are usually used as a stopping medium. For ions with energies ≤ 25 keV/u and a charge state $q \geq 2$, the cross section for one-electron reduction is between 10^{-15} and 10^{-17} cm² [32]. The ions that enter the gas catcher filled with helium at energies < 2 keV/u usually end up thermalized with a charge state of up to 3+ [32].

Once the ions are thermalized, the probability of charge exchange is very low because the first ionization potential of helium is much higher than the second ionization potential of most of the other elements. For example, in the case of tin, the second ionisation potential is 14.633 eV [33], while the first ionization potential for helium is 24.587 eV [33].

The situation is different if there are molecules or other elements present within the buffer gas, introduced on purpose (such as the example of CH₄ in Section 7.3) or as impurities that originate from the buffer gas or outgassing (*e.g.* H₂O, N₂, CO, CO₂ and noble gases such as Ar, Kr, Xe). The ionization potential of molecules M that are usually present as impurities are between 10 eV and 16 eV [32], which enables the following type of reactions

In a helium-gas environment the formation of molecules occurs through an association reaction of an ion R^+ with a ligand molecule M

The ligand molecule M usually originates from impurities. The excited complex R^+M^* can undergo dissociation or it can stabilize by collisions with helium atoms

The ion density n_{R} for thermalized ions inside the buffer gas changes over time in the following way

$$\frac{dn_R}{dt} = -kn_R n_M, \quad (2.31)$$

where n_M is the ligand density and k is the reaction-rate coefficient. In this reaction the time constant is defined as

$$\tau = \frac{1}{kn_M}. \quad (2.32)$$

Therefore, the reaction time for the formation of molecular ions depends on the reaction-rate coefficient and the ligand density. In general, the reaction-rate coefficient that corresponds to the reaction of ions with impurities within a gas catcher is on the order of $k = 1 \cdot 10^{-10} \text{ cm}^3/\text{s}$ [34]. An impurity level of 1 ppm in a gas catcher filled with 50 mbar of helium corresponds to $n_M = 1.35 \cdot 10^{12} \text{ cm}^{-3}$. Thus, the estimated reaction time is 7.4 ms. Chemical reactions between the ions and impurities are often not desired since they lead to ion losses. However, a method to separate isobaric nuclei based on their chemical properties is proposed and discussed in Section 7.3.

The stopping of energetic ions creates ion-electron pairs. Thanks to the applied DC field (see Fig. 2.3), ions and electrons move in opposite directions. Since the mobility of electrons is three orders of magnitude larger than that of ions, the electron density will be significantly lower. This means that recombination of the thermalized ions is less likely to occur. However, whenever ionization occurs to a large extent, an electronic plasma is formed which shields the applied electric field. This may happen if the rate of ions entering the gas catcher is large. In this case, electrons and ions are not pulled apart and reactions between thermalized ions and free electrons will occur

The ion loss dn_i/dt depends on the helium pressure and the electron density

$$\frac{dn_i}{dt} = -\alpha n_i n_e n_{\text{He}}, \quad (2.34)$$

where α is the three-body recombination coefficient and n_i , n_{He} and n_e are the ion, helium and electron densities, respectively. The corresponding time constant τ represents the survival time of the ions and is given as

$$\tau = \frac{1}{\alpha n_{\text{He}} n_e}. \quad (2.35)$$

There is a lack of experimental data for the three-body recombination coefficient for atomic ions. Therefore, it is hard to predict the ion survival time. However, there are indications that the survival time is ≥ 10 ms [32] which is comparable with the ion extraction time.

Additionally, neutralization reactions can occur due to dissociative recombination with electrons. If a molecular ion RM^+ is present in the gas cell, it can be neutralized in reactions with electrons

To avoid ion loss due to recombination, a higher electric field gradient should be applied to separate ions and electrons within the gas catcher and additionally, decrease the extraction time.

2.3 Ion motion in a radio-frequency quadrupole

Radio-Frequency Quadrupoles (RFQs) can be used to extract ions from the gas catcher and transport them downstream for further manipulation. A RFQ consists of four parallel rods (usually round or hyperbolically shaped) which are supplied with a RF voltage. The RF voltage of

two neighboring rods is phase-shifted by 180 degrees compared to the other pair of rods. Some RFQs are additionally supplied with a DC voltage. The electric potential Φ inside a RFQ is given by [35]

$$\Phi = \Phi_0 \frac{x^2 - y^2}{r_0^2}, \quad (2.38)$$

where r_0 is the half of the distance between opposite rods. Φ_0 is the potential difference between the two pairs of rods and it has the following form

$$\Phi_0 = U_{\text{DC}} + V_{\text{RF}} \cos \Omega t, \quad (2.39)$$

where U_{DC} is the DC voltage and V_{RF} is the amplitude of a RF voltage with an angular frequency Ω . The potential distribution in the radial plane for a RFQ is shown in Fig. 2.4 left.

For an ion with mass m and charge q , the equations of motion inside of the RFQ are given by

$$m \frac{d^2 x}{dt^2} = -\frac{2q}{r_0^2} (U_{\text{DC}} + V_{\text{RF}} \cos \Omega t) x, \quad (2.40)$$

$$m \frac{d^2 y}{dt^2} = \frac{2q}{r_0^2} (U_{\text{DC}} + V_{\text{RF}} \cos \Omega t) y. \quad (2.41)$$

By introducing a parameter $\xi = \Omega t/2$, the equations of motion can be modified to

$$\frac{d^2 x}{d\xi^2} = -\frac{8q}{m\Omega^2 r_0^2} (U_{\text{DC}} + V_{\text{RF}} \cos 2\xi) x, \quad (2.42)$$

$$\frac{d^2 y}{d\xi^2} = \frac{8q}{m\Omega^2 r_0^2} (U_{\text{DC}} + V_{\text{RF}} \cos 2\xi) y. \quad (2.43)$$

At this stage, the Mathieu stability parameters $a_{x,y}$ and $q_{x,y}$ can be introduced

$$a_x = -a_y = \frac{8qU_{\text{DC}}}{m\Omega^2 r_0^2}, \quad (2.44)$$

$$q_x = -q_y = \frac{4qV_{\text{RF}}}{m\Omega^2 r_0^2}, \quad (2.45)$$

and both equations 2.42 and 2.43 can be written as

$$\frac{d^2 u}{d\xi^2} - (a_u - 2q_u \cos 2\xi)u = 0, \quad (2.46)$$

Figure 2.4: Left: a transverse cut of the RFQ rods together with the resulting potential distribution. Right: stability diagram of a RFQ. The shaded areas represent Mathieu parameters for which the ion motion is stable in x and y directions, while their intersections indicate parameters for which the ions are radially confined. For practical reasons, usually RFQs are operated in the region of stability closest to $a = q = 0$. Figure adopted from Ref. [36].

where u is either x or y . This equation is known as the Mathieu equation [37] and it describes the ion motion inside of the RFQ. The ion motion is stable only for a combination of a_u and q_u parameters that gives real solutions of the Mathieu equation. The stability diagram defined by these parameters is shown in the right part of Fig. 2.4. The stable ion motion in either x or y directions is indicated by the shaded area, while their intersections show the stability regions. If the combination of a_u and q_u parameters is located within the stability region, the ion motion is stable and the ions can be transported through the RFQ.

In the case when no DC voltage is used ($a_u = 0$), the RFQ operates as an ion guide transmitting a broad range of masses. Otherwise, the RFQ works in a mass-filtering mode where the operating point is in the narrow tip of the stability diagram [38]. This means that for fixed operating parameters of the RFQ ($r_0, V_{\text{RF}}, \Omega, U_{\text{DC}}$) only the mass-over-charge ratio which lies inside of the stability region will be transmitted through the RFQ.

2.4 Penning trap mass measurements

A Penning trap is a device that confines charged particles in three dimensions by using a quadrupolar electric potential in combination with a homogeneous axial magnetic field. This system enables measurements of the cyclotron frequency of the stored particles through which it is possible to determine their mass.

2.4.1 Ion motion in a Penning trap

When an ion with mass-over-charge ratio m/q and velocity v is moving in a magnetic field pointing along the z -axis $\vec{B} = B_0 \vec{z}$, it experiences the Lorentz force $\vec{F}_L = q[\vec{v} \times \vec{B}]$ in addition to the centrifugal force $\vec{F}_c = mv^2/\vec{r}$. As a result, the ion will be confined radially ($x - y$ plane) and orbit with the so-called *free cyclotron frequency* ν_c which can be derived from the Lorentz and the centrifugal forces as

$$\nu_c = \frac{1}{2\pi} \frac{|q|}{m} B. \quad (2.47)$$

To confine the ion axially, an electrostatic quadrupole potential

$$\phi(r, z) = \frac{C_2}{2} \frac{U_0}{d^2} \left(z^2 - \frac{r_0^2}{2} \right) \quad (2.48)$$

is superimposed on the strong magnetic field, where U_0 is the depth of the potential well applied to the electrode structure to confine the ions and (r, z) are cylindrical coordinates with $r = \sqrt{x^2 + y^2}$. C_2 is a dimensionless coefficient that is associated with the trap geometry and $d^2 = \frac{1}{2}(z_0^2 + r_0^2/2)$ denotes the characteristic trap dimension. A sketch of a hyperboloidal and a cylindrical Penning trap is shown in Fig. 2.5. In both configurations, the Penning trap consists of a ring electrode with radius r_0 and two endcaps which are located at distance z_0 from the trap center. In the case of the cylindrical trap, additional correction electrodes are necessary in order to create a harmonic potential near the trap center.

Figure 2.5: A sketch of a hyperboloidal (left) and a cylindrical (right) Penning trap together with their resulting axial potential created for the axial confinement of the ions. Figure adopted from Ref. [39].

Due to the presence of the quadrupolar electrostatic field and the homogeneous magnetic field, the equation of motion is given by

$$\vec{F} = q(-\vec{\nabla}\phi + \vec{v} \times \vec{B}) = m\ddot{\vec{r}}, \quad (2.49)$$

and its solutions lead to the three independent harmonic motions shown in Fig 2.6. The *axial motion* of the ion is determined by the z -component of Eq. 2.49 and it is independent of the magnetic-field strength. There are two motions in the radial plane: the *modified cyclotron motion* that originates from the free cyclotron motion that is modified by the repulsive radial-field component of the electrostatic potential, and the *magnetron motion* that results from an azimuthal $\vec{E} \times \vec{B}$ force.

Figure 2.6: Illustration of three-dimensional ion motion in a Penning trap and its projection onto the radial plane. An ion performs a motion that can be decomposed into three eigen-motions: axial, modified cyclotron, and magnetron. Figure adapted from Ref. [40].

The eigen-frequencies of the three motions are given by

$$\nu_z = \frac{1}{2\pi} \sqrt{\frac{2qU_0C_2}{md^2}}, \quad (2.50)$$

$$\nu_+ = \frac{1}{2}(\nu_c + \sqrt{\nu_c^2 - 2\nu_z^2}), \quad (2.51)$$

$$\nu_- = \frac{1}{2}(\nu_c - \sqrt{\nu_c^2 - 2\nu_z^2}), \quad (2.52)$$

where ν_z , ν_+ and ν_- are the axial, modified cyclotron and magnetron frequencies, respectively, with the following hierarchy for typical operating conditions

$$\nu_+ \gg \nu_z \gg \nu_-. \quad (2.53)$$

Through a series expansion of Eq. 2.52 it can be shown that the magnetron frequency is, to first order, independent of the particle properties

$$\nu_- = \frac{1}{4\pi} \frac{C_2 U_0}{B d^2}. \quad (2.54)$$

The three eigen-frequencies of an ion in a Penning trap are related by the so-called *invariance theorem* [41]

$$\nu_c^2 = \nu_+^2 + \nu_-^2 + \nu_z^2. \quad (2.55)$$

The following equation is valid in case of an ideal trap

$$\nu_c = \nu_+ + \nu_-, \quad (2.56)$$

while in the presence of electric and magnetic field imperfections, it is still a good approximation for a relative precision of a few 10^{-10} [3].

2.4.2 Ion manipulation and preparation

To perform high-precision mass measurements, the amplitudes of the eigen-motions have to be manipulated. In this section dipolar and quadrupolar excitations are introduced together with a mass-selective buffer-gas cooling technique that can be employed in a preparation trap to decrease the amplitudes of the ion motions before performing mass measurements in a measurement trap.

Dipolar and quadrupolar excitation

The ion motion in a Penning trap can be treated as three independent harmonic motions. Therefore, by introducing an external force to the system, it is possible to manipulate the motional amplitudes individually. This is done by introducing a RF potential to the segmented trap electrodes. In order to increase the amplitude of one eigen-motion, a dipolar excitation with a frequency that corresponds to one of its eigen-frequencies has to be introduced. The dipolar field can be created by applying an alternating potential with an amplitude U_d and a frequency ν_d to two opposing segments of the ring electrode as shown in Fig. 2.7 a, and it can be described by

$$\vec{E}_x = \frac{U_d}{r_0} \cos(2\pi\nu_d t + \phi_d) \vec{e}_x, \quad (2.57)$$

where ϕ_d is a phase, r_0 is the radius of the ring electrode and $\vec{e}_x$ is the unit vector.

Considering that the magnetron motion of the ion is, to first order, mass independent, by applying a radio-frequency signal at the frequency $\nu_d = \nu_-$ to the segmented ring electrode (see Fig. 2.7 a), all ions inside the trap are excited to a larger radius. Since the reduced cyclotron frequency is mass dependent, an excitation at $\nu_d = \nu_+$ can be used to selectively increase the motional amplitude of one ion species and thus remove undesired ion species from the trap.

Depending on the difference in the initial phase between the radio-

Figure 2.7: Radial segmentation of the ring electrode to apply an electromagnetic radio-frequency field. In (a) the generation of a dipolar field is shown while in (b) a quadrupolar one is established. Figure adopted from Ref. [42].

frequency signal and the ion motion, a linear increase of the magnetron radius starts immediately for $\Delta\phi = 0$ or it decreases until zero and then it increases linearly for $\Delta\phi = \pi$ [42]. Figure 2.8 shows the temporal evolution of the magnetron radius for different initial phase differences as a function of the excitation time.

A quadrupolar excitation enables coupling and conversion between two eigen-motions, for example if an excitation at the sum of their eigen-frequencies, $\nu_+ + \nu_-$, is applied. A quadrupolar field can be created by applying an alternating potential to a four-fold segmented ring electrode such that two opposite segments have the same phase as shown in Fig. 2.7 b. The quadrupolar field can be described by

$$\vec{E}_x = \frac{2U_q}{r_0^2} \cdot \cos(2\pi\nu_q t + \phi_q) y \vec{e}_x, \quad (2.58)$$

$$\vec{E}_y = \frac{2U_q}{r_0^2} \cdot \cos(2\pi\nu_q t + \phi_q) x \vec{e}_y, \quad (2.59)$$

Figure 2.8: The evolution of the magnetron radius as a function of the dipolar excitation time for different initial phase differences ($\Delta\phi = 0, \pi/2, \pi$) between the applied dipole signal and the ion motion. Figure adopted from Ref. [42].

where U_q is the amplitude of the excitation signal with a frequency ν_q and an initial phase ϕ_q at the radius r_0 . A full conversion from one radial motion into the other is achieved if the excitation frequency corresponds to the sum of magnetron and reduced cyclotron frequencies

$$\nu_q = \nu_- + \nu_+ = \nu_c. \quad (2.60)$$

In that case, a continuous conversion between the magnetron and modified cyclotron motions takes place. The time needed for one full conversion of one motion into the other one, T_{conv} , is given by [2]

$$T_{\text{conv}} = \frac{4\pi r_0^2 B}{U_q}. \quad (2.61)$$

The conversion process from the magnetron motion into the modified cyclotron motion is shown in Fig. 2.9. The circle around the origin represents the initial magnetron motion. When a quadrupolar excitation at the resonance frequency $\nu_q = \nu_c$ is applied, the magnetron radius starts to reduce, and the modified cyclotron radius increases. After the time T_{conv} , the magnetron radius has been decreased to zero, while the modified cyclotron radius has the magnitude of the initial magnetron radius. For a non-resonant excitation ($\nu_q \neq \nu_c$) a partial conversion will occur.

Figure 2.9: The ion trajectory during conversion from a pure magnetron motion into a pure modified cyclotron motion. The first half of the conversion is shown in (a) and the second half of the conversion process is shown in (b). Figure adopted from Ref. [2].

Buffer-gas cooling technique

A cooling technique developed to reduce the axial and reduced cyclotron motion of ions stored in a Penning trap exploits collisions with a buffer gas, usually helium. Due to low collision energies, the process can be

described by a viscous-drag model according to which the ion with mass m and velocity $\vec{v}$ experiences a damping force $\vec{F} = -\delta m \vec{v}$, where δ is connected to the ion mobility K (see Eqs. 2.13-2.15). In the presence of the damping force, the axial and the radial eigen-motions will exponentially lose energy. The change in the initial radius of the radial motion $R_{\pm}(0)$ can be described as [43]

$$R_{\pm}(t) = R_{\pm}(0) e^{\mp \delta \frac{\nu_{\pm}}{\nu_{+} - \nu_{-}} t}. \quad (2.62)$$

The radius of the modified cyclotron motion R_{+} will decrease over time while the radius of the magnetron motion R_{-} will increase. As the modified cyclotron frequency ν_{+} is much higher than the magnetron frequency ν_{-} , the modified cyclotron motion is damped faster (see Fig. 2.10 a). In order to avoid ion loss due to a constantly increasing magnetron radius, a quadrupolar excitation at the free cyclotron frequency of the ion of interest ν_c is applied, which leads to centering of the ion in the Penning trap (see Fig. 2.10 b).

As ν_c depends on the mass-over-charge ratio of an ion, the buffer-gas cooling technique is mass-selective [43]. For example, at the SHIPTRAP setup (see Chapter 3), the buffer-gas cooling technique allows isobaric purification of the species of interest with a mass resolving power, defined as $\nu_c/\delta\nu_c$, of about 100,000 [5].

The spatial radial distribution of the ion cloud is described by a probability density function. Approximated by a Gaussian distribution, the full width at a half maximum W is given by [3]

$$W \approx \frac{2.35}{\pi \nu_{+}} \sqrt{\frac{k_B T}{2m}}, \quad (2.63)$$

where m is the mass of the ion and T is the temperature of the buffer-gas. For singly-charged ions of ^{133}Cs in a 7 T magnetic field at room temperature, the radial distribution of the cooled ion cloud is about $90 \mu\text{m}$.

Figure 2.10: An illustration of the radial ion motion during buffer-gas cooling in a Penning trap. (a) In the absence of RF fields, the cyclotron radius is reduced very quickly while the magnetron radius increases slowly. (b) By introducing a quadrupolar excitation at $\nu_{RF} = \nu_c$, the two radial motions are coupled leading to a slow decrease of the cyclotron and the magnetron radius. Figure adopted from Ref. [2].

2.4.3 Phase-Imaging Ion-Cyclotron-Resonance Technique

One of the recent techniques developed to determine the cyclotron frequency of a charged particle inside a Penning trap is the Phase-Imaging Ion-Cyclotron-Resonance Technique (PI-ICR) [3]. It is based on the projection of the phase of the radial ion motion in the trap after a given accumulation time on a position-sensitive detector. Compared to the ion motion inside of the trap, the projection onto the detector is magnified with a magnification factor G , which in absence of an electric field is given by [3]

$$G \approx \sqrt{\frac{B_z^{\text{trap}}}{B_z^{\text{det}}}}, \quad (2.64)$$

where B_z^{trap} and B_z^{det} are the z components of the magnetic field at the position of the trap-center and the detector.

From the projected phases, it is possible to obtain the magnetron ν_- and modified cyclotron ν_+ frequency, whose sum according to Eq. 2.56 determines the free cyclotron frequency ν_c . Two excitation patterns are used in the PI-ICR technique: a single pattern for an independent measurements of ν_- and ν_+ and a double pattern for a direct measurement of ν_c . In the following, both patterns will be introduced based on Ref. [3].

Single pattern: independent measurements of ν_- and ν_+

Suppose that the ions in the trap are well cooled and centered. When a dipolar excitation is applied at the frequency ν_- or ν_+ the ions are radially excited to an average radius $r_{\pm}$ and the reference phase is set. After a given time t of excitation-free propagation, the ions will reach the final phase (see Fig. 2.11) and thus build up a total phase of

$$\phi_{\pm} + 2\pi n_{\pm} = 2\pi\nu_{\pm}t, \quad (2.65)$$

where $\phi_{\pm}$ is the polar angle between reference and final phases, $n_{\pm}$ is the number of full revolutions and $\nu_{\pm}$ represents radial frequency.

The excitation-pulse scheme for the measurement of the magnetron and modified cyclotron frequencies is shown in Fig. 2.12. In the case of a magnetron frequency measurement, cooled ions are injected into the measurement trap and subsequently excited by the dipolar RF-pulse. If the ions are ejected from the trap right after the dipolar excitation, the reference phase is recorded at the detector. To record the final phase, the ejection of ions is done after a phase-evolution time t . Therefore, the total phase is $\phi_- + 2\pi n_- = 2\pi\nu_-t$.

Compared to ν_- , the measurement of the modified cyclotron frequency contains additional steps. Step 1a is introduced to damp the initial magnetron motion and in that way avoid a possible systematical shift of the ν_+ frequency. After a dipolar excitation of the modified cyclotron

Figure 2.11: The radial ion motion inside a Penning trap projected onto a position-sensitive detector. The spot that corresponds to the trap-center is obtained without applying any excitation, the reference phase is a projection of the radially excited ion motion, while the final phase corresponds to the same motion that is projected after building up a certain phase during time t . Figure adopted from Ref. [44].

Figure 2.12: Excitation-pulse scheme for the measurement of the magnetron (left) and modified cyclotron (right) frequencies. For details, see text.

motion, the fast cyclotron motion is converted into the slow magnetron motion by applying a quadrupolar RF-pulse at the cyclotron frequency. This is done in step 2a with the aim to avoid substantial angular spread on the detector due to the time-of-flight distribution of the ions which is related to the ejection from different axial positions due to the axial motion which cannot be damped completely. Here, it is important to note that the phase between the reference and final phase of the modified cyclotron frequency after conversion preserves the modulus of the angle but flips the sign. If the conversion is done immediately after the dipolar excitation and the ions are ejected to the detector, then the reference phase is recorded. In order to project the final phase, it is necessary to delay the conversion for a phase-evolution time t . The total phase is $\phi_+ + 2\pi n_+ = 2\pi\nu_+ t$.

The projection of the trap-center is done in the same way as the projection of the magnetron motion but without any excitation applied. In general, the position of the trap-center and the reference phase are stable over extended periods and are, therefore, usually measured once per day [3]. Finally, once the radial frequencies are determined from the total phases, the cyclotron frequency is obtained as their sum $\nu_c = \nu_- + \nu_+$.

Double pattern: a direct measurement of ν_c

A double-pattern scheme enables a direct measurement of the free cyclotron frequency ν_c by measuring only the final phases of the magnetron ϕ_- and modified cyclotron motion ϕ_+ , the so-called magnetron and cyclotron spots. The excitation-pulse scheme is shown in Fig. 2.13. The main difference between the two patterns is in step 4. To project the magnetron spot, the conversion has to be done immediately after the dipole excitation so that the ions perform magnetron motion during accumulation time t . For the projection of the modified cyclotron spot, the ions perform the modified cyclotron motion during accumulation time. The conversion pulse is applied just before the ejection from the trap. The projection of the magnetron and the modified cyclotron spots are shown in Fig. 2.14.

Figure 2.13: Excitation-pulse scheme for the direct measurement of free cyclotron frequency. For details, see text.

Figure 2.14: The magnetron and the modified cyclotron motion in a Penning trap projected onto a position-sensitive detector after a phase accumulation time t . For more details see text. Figure adopted from Ref. [44].

The relative phases $\phi_{\pm}$ of radial motions built up during the time t are given by

$$\phi_{\pm} = \pm(2\pi n_{\pm} - 2\pi\nu_{\pm}t) + \phi_0, \quad (2.66)$$

where ϕ_0 represents the initial phase. By summing up the relative phases ϕ_{-} and ϕ_{+} , it is possible to calculate the free cyclotron frequency

$$\nu_c = \frac{\phi + 2\pi(n_- + n_+)}{2\pi t}, \quad (2.67)$$

where $\phi = \phi_- - \phi_+$. The two phases, ϕ_+ and ϕ_- have opposite signs due to the conversion of the modified cyclotron into magnetron motion.

2.4.4 Imperfections of a real Penning trap

The above-mentioned equations are valid in the case of a single ion confined in an ideal Penning trap, where all eigen-frequencies are totally decoupled and depend only on the mass-over-charge m/q of the stored ion, the electric and the magnetic field strengths, and the dimension of the trap. In the case of a real Penning trap, there are field inhomogeneities that are a consequence of a non-ideal trap geometry, field instabilities, axes misalignments and other imperfections, which lead to systematic shifts of the ion frequencies. In the following, the most common sources of such systematic shifts are introduced.

Imperfections of the electrostatic field

One of the sources of electrostatic anharmonicities originates from the trap geometry. In a real trap, the electrodes have a finite length and they are segmented for ion manipulation. Additionally, machining imperfections, misalignment of the trap electrodes or electrostatic potential offsets may be present. This implies that the electrostatic potential is never purely quadrupolar. As a consequence, the harmonic eigenmotions become coupled leading to the radial frequency shift [45]

$$\begin{aligned} \Delta\nu_{\pm} = & \mp \frac{C_4}{C_2} \frac{3}{4d^2} \frac{\nu_+\nu_-}{\nu_+ - \nu_-} (2z^2 - r_{\pm}^2 - r_{\mp}^2) \mp \\ & \mp \frac{C_6}{C_2} \frac{15}{8d^4} \frac{\nu_+\nu_-}{\nu_+ - \nu_-} (3z^4 - r_{\pm}^4 + 3r_{\mp}^4 - 6r_{\pm}^2 z^2 - 12r_{\mp}^2 z^2 + 6r_+^2 r_-^2). \end{aligned} \quad (2.68)$$

The dimensionless coefficients C_2 , C_4 and C_6 are functions of the electrode geometry and the applied voltages, z is the amplitude of the axial

motion, while r_- and r_+ represent the radius of the magnetron and modified cyclotron motions, respectively. The resulting shift of the free cyclotron frequency is $\Delta\nu_c = \Delta\nu_- + \Delta\nu_+$.

Imperfections of the magnetic field

The intrinsic spatial homogeneity of the magnetic field of superconducting magnets that are used in Penning trap measurements is on the order of $\Delta B/B = 10^{-7}$ in a volume of 1 cm^3 . However, the homogeneity of the magnetic field in the trap center can be further perturbed due to the use of materials with a non-zero magnetic susceptibility since in the strong magnetic field these materials will magnetize. The absolute shift in the free cyclotron frequency due to higher-order coefficients of the magnetic field is [41]

$$\Delta\nu_c \approx \beta_2\nu_c(r_z^2 - r_-^2), \quad (2.69)$$

where β_2 denotes the relative strength of the hexapole component of the magnetic field.

The magnetic field of a superconducting magnet decreases steadily over time, which is known as flux creep [46]. For long periods of up to years, it can be approximated by a linear function. Furthermore, the magnetic permeability of the materials that surround the superconducting coil depends on the temperature. Therefore, temperature changes within the magnet liquid-helium cryostat can result in magnetic field fluctuations over time. These fluctuations of the magnetic field can be minimized by stabilizing the temperature of the magnet bore and the pressure inside the cryostat.

Misalignment

A misalignment of the electric-field axis with respect to the magnetic-field axis results in a coupling of the eigen-motions and thus shifts the free cyclotron frequency. The free cyclotron frequency shift can be approximated as [47]

$$\Delta\nu_c \approx \frac{9}{4}\nu_- \sin^2\theta, \quad (2.70)$$

where θ represents the angle of the tilt. To first order, this shift is mass independent. It can be estimated that for a relative shift of the free cyclotron frequency below 10^{-9} , the tilt angle has to be less than 0.05° .

When employing the PI-ICR technique, an inclination of the detector with respect to the symmetry axis of the trap electrodes results, furthermore, in a distortion of the orbit-image projection. The projected image has the shape of an ellipse which causes a discrepancy between the actual phase ϕ and measured angle ϕ^* . The shift of the radial-motion frequency caused by the distortion of the orbit image is given by [3]

$$\begin{aligned} \Delta\nu &= \frac{\phi - \phi^*}{2\pi t} \\ &= \frac{\phi - [\arctan(\tan(\beta + \phi)/\cos\delta) - \arctan(\tan\beta/\cos\delta)]}{2\pi t}, \end{aligned} \quad (2.71)$$

where t is the accumulation time, β is the polar angle between the minor axis of the ellipse and the line passing through the origin of the ellipse and the magnetron spot, and δ is inclination angle which can be obtained from the ratio of the minor to the major radius of ellipse ($r_{\min}/r_{\max} = \cos\delta$) that is projected on the detector.

Storage of more than one ion

If there are ions with different masses simultaneously stored in a Penning trap, their center-of-mass motions will interact leading to a cyclotron frequency shift [48]. In the case when the same ion species are stored, this effect is not observed. However, if there is a large ion cloud stored inside a Penning trap, the resulting space charge will create an additional electric field. This field can introduce inhomogeneities

that affect the ion motion leading to the frequency shift. A procedure called “ion-number class” analysis is developed in order to correct the cyclotron frequency shift [49].

2.4.5 Mass-determination principle

According to Eq. 2.47, the mass m of an ion is inversely proportional to the cyclotron frequency ν_c . Therefore, by measuring the cyclotron frequency of an ion, ν_c , with a high accuracy, it is possible to determine its mass accurately. The condition for this is that the magnetic field B is known with comparable accuracy as well. This is achieved by measuring the cyclotron frequency of a reference ion, ν_{ref} , with well-known atomic mass m_{ref} and charge q_{ref} . Considering that the cyclotron frequency of an ion of interest measured at time t_1 is given as

$$\nu_c(t_1) = \frac{1}{2\pi} \frac{q}{m} \cdot B(t_1) \quad (2.72)$$

and the cyclotron frequency of the reference ion measured at time t_2 is

$$\nu_c^{\text{ref}}(t_2) = \frac{1}{2\pi} \frac{q_{\text{ref}}}{m_{\text{ref}}} \cdot B(t_2), \quad (2.73)$$

then the mass ratio can be derived as

$$\frac{m}{m_{\text{ref}}} = \frac{q}{q_{\text{ref}}} \cdot \frac{\nu_c^{\text{ref}}(t_2)}{\nu_c(t_1)} \cdot \frac{B(t_1)}{B(t_2)}. \quad (2.74)$$

Assuming that the ion of interest and the reference ion are measured at the same time ($t_1 = t_2$), the magnetic-field ratio cancels out and the relation simplifies to

$$\frac{m}{m_{\text{ref}}} = \frac{q}{q_{\text{ref}}} \cdot \frac{\nu_c^{\text{ref}}}{\nu_c} = \frac{q}{q_{\text{ref}}} \cdot R, \quad (2.75)$$

where R is called a cyclotron frequency ratio. As discussed in Section 2.4.4, the magnetic field changes over time and for Eq. 2.75 to be valid, either a linear or polynomial interpolation method can be applied to obtain the reference frequency at the time when the ion of interest is measured (for details, see Section 5.3).

The mass of the ion of interest can be determined as

$$m = R \frac{q}{q_{\text{ref}}} (m_{\text{ref}} - q_{\text{ref}} m_e) + q m_e, \quad (2.76)$$

where m_e is the mass of the electron.

The SHIPTRAP facility

The SHIPTRAP high-precision mass spectrometer is located behind the velocity filter SHIP (Separator for Heavy Ion reaction Products) [50] at the GSI Helmholtz Centre for Heavy Ion Research, Darmstadt, Germany. The fusion-evaporation products separated by SHIP (see Fig. 3.1) are injected into the cryogenic gas-stopping cell where they are slowed down and thermalized to enable low-energy experiments. From there, the ions are extracted and transferred to a RFQ buncher where a continuous ion beam is converted into ion bunches. The cooled ion bunches are then transported to a double Penning-trap system where the ion motion is manipulated and finally projected onto a detector for high-precision mass measurements.

In this chapter, the velocity filter SHIP and the major components of the current SHIPTRAP setup shown in Fig. 3.2 will be described. In the future, a new gas catcher complementary to the cryogenic gas-stopping cell will be used to study particular ion species for which isobaric separation is needed. This gas catcher is built and its technical details are given in Chapter 8.

Figure 3.1: Schematic view of the velocity filter SHIP. The primary beam (orange) hits a target wheel and it is directed to the beam stop together with undesired reaction products (red). The fusion-evaporation products are filtered according to their velocity by a special combination of electric and magnetic fields and transmitted towards SHIPTRAP.

Figure 3.2: Schematic view of the SHIPTRAP setup and its major components (modified from Ref. [51]): the cryogenic-gas stopping cell, the extraction RFQ and the buncher for ion preparation, the reference ion sources and the Penning traps. The triangles indicate detector positions.

3.1 The velocity filter SHIP

The velocity filter SHIP was built for in-flight separation and studies of superheavy isotopes produced in fusion-evaporation reactions [50]. It is well-known for the discovery of the elements with proton number $Z = 107 - 112$ [52–56]. Figure 3.1 illustrates the electrostatic and magnetic components of the separator.

The UNiversal Linear ACcelerator (UNILAC) [57] delivers pulsed heavy-ion beams (orange line in Fig. 3.1) at intensities of typically $6 \cdot 10^{12}$ particles per second towards SHIP located at the zero degree beam line where the beam hits a rotating target wheel. The beam energies are around 5 MeV/u, which is sufficient to overcome the Coulomb barrier between the projectile and the target nucleus. The wheel contains 8 target segments with a thickness of about $500 \mu\text{g}/\text{cm}^2$ and it rotates with a frequency of 1125 turns per minute which is matched to the pulsed-beam structure of the UNILAC. The fusion-evaporation residues that leave the target (yellow line in Fig. 3.1) are kinematically separated from the primary beam and other reaction products (red line) by applying a certain combination of electrostatic and Lorentz forces, $F_e = qE$ and $F_m = qvB$, respectively. The electric and magnetic fields are perpendicular to each other and to the beam axis, which allows only ions that have a certain velocity $v = E/B$ to be transmitted through the separator. The electromagnetic quadrupole triplets at the beginning and the end of the separator are used for focusing. The last magnetic dipole bends the beam up to 7.5° to reduce the transmission of scattered primary beam and unwanted reaction products. The resulting transmission efficiency depends on the reaction and it is in the range of 20% to 50% [58].

3.2 The cryogenic gas cell and extraction RFQ

The reaction products delivered by SHIP, with kinetic energies on the order of several tens of MeV, pass through optional degrader foils and enter the Cryogenic Gas Cell (CGC) through a few μm -thin Ti window foil. The Ti foil thickness depends on the energy of the incoming ion

beam and it is optimized by using the TRIM code [18] in order to achieve maximum stopping efficiency inside the CGC. The ions continue losing their energy through collisions with the He buffer gas inside the CGC, typically at a temperature of 40 K and at densities that correspond to room-temperature pressures of about 50 mbar (see Fig. 3.2). The chamber has a length of 450 mm and a diameter of 400 mm and it is cooled by a cryocooler. In order to provide good thermal insulation, it is placed inside an evacuated chamber that contains multi-layer insulation foils. In general, an important requirement for gas cells is to reduce the amount of impurities and thus prevent a decrease in the efficiency due to unwanted molecular-ion formation, recombination and charge-exchange reactions. For this reason, ultra-high vacuum conditions are required before filling the CGC with highly-purified He gas. The advantage of cryogenic operation is that the remaining impurities are frozen out at the surfaces.

To extract the stopped ions out of the gas cell, a DC voltage gradient is applied to the 8 cylindrical electrodes of the DC cage that guide the ions towards the RF funnel consisting of 76 ring electrodes with successively reduced diameter. The RF funnel focuses the ions towards the de-Laval nozzle that connects the high-pressure area of the CGC with the low-pressure area ($\sim 10^{-2}$ mbar) of the extraction RFQ. In this region, the ions are extracted by the gas flow which is optimized to achieve a supersonic gas jet for an efficient injection into the extraction RFQ. The RFQ rods are segmented in order to apply a DC gradient that guides the ions to the RFQ buncher. The physical processes that occur inside the gas catcher are described in Section 2.2 and a detailed description of the CGC and its performance can be found in Refs. [23, 51].

3.3 The RFQ buncher

To efficiently capture the ions in the Penning trap, it is necessary to convert a continuous ion beam into ion bunches with minimum energy and time spread. Therefore, the ions are transported from the extraction RFQ to a RFQ cooler and buncher. The DC voltage, applied to

the segmented RFQ rods, gradually decreases and creates a potential minimum at the second to last segment.

The RFQ buncher is operated at a helium pressure of around $5 \cdot 10^{-3}$ mbar. While moving through the RFQ buncher, the ions lose their kinetic energy in collisions with helium atoms and are then stored at the potential minimum. Here, the ions are collected for tens or hundreds of milliseconds before being ejected by lowering the potential of the last segment. Similar to the case of the CGC, the gas purity plays an important role in order to prevent charge-exchange reactions and thus, special care is taken to ensure ultra-high vacuum conditions as well as to purify the He gas. More details about the RFQ buncher and its performance can be found in Ref. [59].

3.4 The Penning trap system

As shown in Fig. 3.2, the cooled ion bunches are injected into the double Penning-trap system located inside a 7 T superconducting magnet. The trap electrodes are cylindrical and made out of gold-plated oxygen-free high conductivity (OFHC) copper, while alumina (aluminum oxide) is used for electric insulation. These materials were chosen in order to minimize the distortion of the magnetic field and enable ultra-high vacuum conditions.

Figure 3.3 shows the double Penning-trap system. The first trap is called the Preparation Trap (PT) and it allows isobaric purification of the species of interest with a mass resolving power of about 100,000 using the side-band cooling technique (see Section 2.4.2). It is 212 mm long and contains an eight-fold segmented ring electrode, a pair of two-fold segmented correction electrodes, a pair of correction electrodes without segmentation and three pairs of endcap electrodes. One of the endcap electrodes contains an inlet for helium gas in order to enable buffer gas cooling at a pressure of around $5 \cdot 10^{-5}$ mbar.

The ions of interest are then transferred into the Measurement Trap (MT) where their radial motion is excited and its frequencies can be

Figure 3.3: Schematic view of the Penning trap system at SHIPTRAP.

measured. The MT is 185 mm long and consists of an eight-fold segmented ring electrode, one pair of two-fold segmented correction electrodes and three pairs of endcap electrodes.

The traps are separated by a 52 mm-long diaphragm with a diameter of 1 mm. It consists of the last endcap electrode of the PT and the first endcap electrode of the MT and acts as a pumping barrier. For more details about the design of the trap system see Refs. [60, 61].

3.5 The reference-ion sources

The SHIPTRAP setup is equipped with two surface ion sources as well as with a laser-ablation ion source which provide reference ions of various elements and with different mass-to-charge ratios. As shown in Fig. 3.2, the sources are installed in off-axis extensions of the main beamline and the delivered ions are efficiently transported to the Pen-

ning traps via an electrostatic quadrupole bender. The ions are produced at stable count rates and used either as reference ions for magnetic field calibration or for studies of systematic uncertainties.

Additionally, the laser-ablation ion source has been used in the high-precision off-line measurements with stable and long-lived ions [62, 63]. It exploits a frequency-doubled Nd:YAG laser at 532 nm to irradiate a small target sample and produces ions. The target consists of a thin metallic foil or a liquid-solution deposited and dried on a titanium supporting foil. The target holder can accommodate multiple samples and is driven by a stepper motor that allows fine rotation in order to scan the position of the same sample as well as to access another one. Once ablated from the surface of the sample, the ions are injected into a mini-RFQ, which consists of an injection electrode, a RFQ and an ejection electrode [64]. Here, it is possible to confine the ions axially by applying electrostatic voltages to the injection electrode, the rods and the ejection electrode in a way that a potential minimum is created in the mini-RFQ volume. Through collisions with helium buffer-gas atoms at a pressure around 0.01 mbar, the ions are thermalized and accumulated in the potential minimum inside the RFQ structure. By lowering the bias potential of the ejection electrode, a bunch of cooled ions is ejected, accelerated and guided towards the Penning traps. In order to increase the sensitivity for offline experiments on rare isotopes, simulation studies of the laser-ablation ion source were performed and details can be found in Ref. [65].

3.6 The detection system

At SHIPTRAP, there are three detection points indicated as triangles in Fig. 3.2. Movable feedthroughs that contain a silicon detector and a micro-channel plate (MCP) or a channeltron detector (see Fig. 3.4) are installed at the positions D1 and D2 and allow to replace ion optics elements with one of the above-mentioned detectors. These detectors are used to monitor and optimize the ion transmission efficiency. A delay-line position-sensitive MCP detector is installed at the position

Figure 3.4: Detection systems used at SHIPTRAP. For details, see text.

D3 to record the ion's radial position and its Time-of-Flight (ToF) after ejection from the trap system.

3.6.1 Silicon detector

The silicon detectors are used to record an α spectrum from radioactive ions transmitted during an experiment. The radioactive ions are deposited on an aluminum foil with a thickness of $0.5\ \mu\text{m}$ (see Fig. 3.4). The emitted α particles hit the detector installed right behind the foil and produce electron-hole pairs. The electron-hole pairs can be collected resulting in an electrical signal proportional to the energy of the α particle. This allows identifying radioactive isotopes and monitoring the efficiency of the setup.

3.6.2 MCP and channeltron detector

MCP and channeltron detectors are used to record ToF spectra from which it is possible to identify ions by their mass-over-charge ratio. This

is important for tuning the beamline, as well as to monitor the purity of the system. The MCP detector is shown in Fig. 3.4. The main difference between the MCP and the channeltron detectors is that the MCP contains an assembly of small channels while the channeltron has only one channel. However, the working principle for both detectors is the same. When an ion hits the detector surface, it produces few electrons that are guided by a potential gradient through the detector channel(s). The internal walls of the channel are covered with metallic materials from which secondary electrons can be easily emitted leading to a detectable current signal at the anode.

3.6.3 Delay-line position-sensitive MCP detector

A delay-line position-sensitive MCP detector (DLD40 from RoentDek GmbH) [66] is installed behind the traps to project the radial ion motion from which the mass can be determined with high accuracy. As shown in Fig. 3.4, it consists of two MCPs and the delay-line anode. The active area of the MCPs is 42 mm in diameter. The MCP signal can be used to record the ToF of the ejected ions. A two-dimensional position determination can be obtained by the delay-line anode that contains two layers of wire pairs wound in x and y directions. One wire from each layer is called signal wire and it is biased to a slightly more positive voltage than the other wire that is called the reference wire. As a result, the electron cascade produced by the MCPs is mostly collected by the signal wire of both layers. The signal propagates to the ends of each wire with a speed close to the speed of light. From the difference in the signal arrival time at both ends of signal and reference wire, for each layer independently, it is possible to obtain the positions in x and y directions. The sums of the signal arrival times have an intrinsic distribution for which a calibration is done. This calibration is used in order to filter true events from the background. The detector has a position resolution of $70\ \mu\text{m}$, a time resolution of 10 ns and an efficiency of around 30%.

Towards high-precision mass measurements of super-heavy elements

Experimental investigations of the elements with the highest atomic numbers provide valuable information on their nuclear properties and the limit of nuclear existence. These elements are so far only accessible through fusion-evaporation reactions at accelerator-based facilities. The low production rates and relatively short half-lives represent the main challenges in their study. The SHIPTRAP mass spectrometer described in Chapter 3 has been built and optimized for high-precision mass measurements of heavy and superheavy elements. First pioneering experiments performed in this region provided important input to benchmark and improve theoretical models [5, 6, 67]. This chapter describes the motivation for extending the experimental program towards heavier species and gives an overview of the SHIPTRAP experimental campaign performed in 2018.

4.1 Exploring the superheavy elements

The nuclei with the highest numbers of protons and neutrons represent a unique testing ground to explore the nuclear structure at the extremes of nuclear stability. In such heavy systems, there is a complex interplay between the repulsive Coulomb force among protons that tends to disintegrate the nucleus and the nuclear force that holds its constituents together. According to the liquid drop model, in nuclei with a proton number larger than $Z \approx 100$ the repulsive force is so strong that they would decay immediately by spontaneous fission [68]. The model is based on a macroscopic approach in which the atomic nucleus is considered to be a drop of incompressible nuclear matter kept together by the nuclear force [69]. While it predicts well the main trends in atomic masses, it does not reproduce the relatively high stability for certain proton and neutron numbers, as well as the existence of the heaviest elements with atomic numbers up to $Z = 118$. To explain the increased stability for so-called magic numbers of protons and neutrons, the nuclear shell model was introduced [70,71]. According to the shell model, the nucleons are arranged in shells and enhanced stability is observed when these shells are closed. Thus, superheavy elements ($Z \geq 104$) exist only because of enhanced nuclear stability due to shell effects. Besides, theoretical models predict the existence of an island of stability where long-lived spherical nuclei with closed-shell configuration would be located. The predictions have, still, not converged to the same location of the island of stability (potential magic numbers are predicted to be at $Z = 114, 120$ or 126 and at $N = 172$ or $N = 184$) [72] and an experimental verification at present is not technically feasible.

However, by studying nuclear properties of lighter elements, such as nobelium and lawrencium ($Z = 102$ and $Z = 103$, respectively), it is possible to indirectly obtain valuable information about heavier elements and provide input to improve theoretical predictions concerning the location of the island of stability. At GSI, Germany, these elements are being investigated utilizing Penning-trap mass spectrometry, laser spectroscopy and decay spectroscopy.

High-precision mass measurements provide absolute mass values and model-independent binding energies that reflect nuclear shell effects. Direct Penning trap mass spectrometry of $^{252-255}\text{No}$ and $^{255,256}\text{Lr}$ performed at SHIPTRAP allowed mapping the strength of the deformed neutron shell closure at $N = 152$ [6, 7]. Furthermore, these measurements in combination with decay spectroscopy studies improved indirectly the values of the masses of heavier nuclides connected to them via α -decay chains [7].

Some nuclei in this region have excited metastable states called *K*-isomers. *K* represents the total spin projection onto the symmetry axis in axially symmetric nuclei. The existence of *K* isomers occurs for a large difference in *K* quantum number between the isomeric state and the ground state. For an excited state with a large *K* value, the decay to a ground state with lower *K* requires a large change in *K* quantum number. Emission of radiation with high multipolarity is usually strongly hindered which gives rise to *K*-isomers. Both *K*-isomers as well as spin isomers with low excitation energies decay usually by internal conversion that is accompanied by the emission of low-energy conversion electrons. Here, decay spectroscopy studies may be difficult due to possible summing in the energy of electrons coming from the excited state and emitted particles originating from the decay of the corresponding ground state. Thanks to the high mass-resolving power of the PI-ICR technique (see Section 2.4.3), Penning-trap mass spectrometry can provide a powerful alternative to determine the excitation energy of low-lying isomeric states.

Performing experimental research in this region is still a challenging task since the number of available nuclei is limited by low yields. As an example, in the previous experimental campaign at SHIPTRAP, it took 93 hours to detect 48 events of ^{256}Lr whose production cross section is 60 nb [6]. To measure nuclei with even lower rates, the performance of the SHIPTRAP setup had to be improved. Therefore, the cryogenic gas-stopping cell (see Section 3.2) was implemented and optimized to increase the overall efficiency of the setup [23, 51]. The development of the PI-ICR technique enabled a forty-fold increase in the resolving

power and a 5-fold gain in precision compared to the Time-Of-Flight Ion-Cyclotron-Resonance (TOF-ICR) technique used in the previous experimental campaign [3]. Thanks to these improvements, during the SHIPTRAP experimental campaign in 2018 (see the next section) it was possible to access nuclei with cross section as low as 15 nb and to measure isomeric states with excitation energies down to 30 keV. This success proved the capability of the SHIPTRAP mass spectrometer to access the superheavy elements and deliver results crucial to explore this region.

4.2 Online measurement campaign

During June and July in 2018, within the FAIR Phase-0 program, the masses of ground states of $^{251,254}\text{No}$ and $^{254-256}\text{Lr}$ and excited states of $^{251\text{m},254\text{m}}\text{No}$ and $^{254\text{m},255\text{m}}\text{Lr}$ were measured at the SHIPTRAP mass spectrometer. In addition, the first direct mass spectrometry of a super-heavy element ^{257}Rf was performed. This section provides a description of the experimental campaign and the measurement process.

4.2.1 Production of nobelium, lawrencium and rutherfordium isotopes

As discussed in Section 2.1, the production cross section depends on the choice of projectile and target, as well as on the projectile energy. Therefore, the fusion-evaporation reactions and the primary-beam energies are chosen to obtain the maximum yields. Table 4.1 shows the reactions together with the corresponding primary-beam energies that were used to produce isotopes of nobelium, lawrencium and rutherfordium at the SHIP separator.

A $^{48}\text{Ca}^{10+}$ beam from UNILAC [57] was used for the production of nobelium and lawrencium isotopes. The second block of measurements was performed by using a $^{50}\text{Ti}^{12+}$ beam in order to produce ^{257}Rf . In both cases, the beam was delivered to the SHIP separator in pulses of 5 ms at a repetition rate of 50 Hz. While the calcium beam was delivered

Table 4.1: Fusion-evaporation reactions and primary beam energies used for the production of nobelium, lawrencium and rutherfordium isotopes. The production cross sections comprise of isomeric states. The target thickness is about $450 \mu\text{g}/\text{cm}^2$.

Isotope	Reaction	Beam energy [MeV/u]	Cross section [nb]
$^{251\text{g,m}}\text{No}$	$^{206}\text{Pb}(^{48}\text{Ca}, 3\text{n})$	4.80	15 [73]
$^{254\text{g,m}}\text{No}$	$^{208}\text{Pb}(^{48}\text{Ca}, 2\text{n})$	4.56	1800 [7]
$^{254\text{g,m}}\text{Lr}$	$^{209}\text{Bi}(^{48}\text{Ca}, 3\text{n})$	4.81	22 [74]
$^{255\text{g,m}}\text{Lr}$	$^{209}\text{Bi}(^{48}\text{Ca}, 2\text{n})$	4.56	250 [75]
$^{256\text{g}}\text{Lr}$	$^{209}\text{Bi}(^{48}\text{Ca}, \text{n})$	4.50	60 [74]
$^{257\text{g}}\text{Rf}$	$^{208}\text{Pb}(^{50}\text{Ti}, \text{n})$	4.65	15 [76]

by the UNILAC with an average intensity of 1 particle μA on target, the titanium beam reached an average intensity of about one quarter compared to the calcium beam due to tests related to the ion source which were performed in order to identify the most suitable production scheme.

PbS targets were used for the production of nobelium and rutherfordium isotopes. These targets were enriched (99.9%) in one particular isotope (^{206}Pb or ^{208}Pb) and had to be exchanged during the experimental campaign according to the desired reactions. For the production of lawrencium isotopes, Bi_2O_3 targets were utilized. In order to ensure the highest production, the targets were continuously monitored and periodically exchanged when their surface was degraded due to radiation damage.

4.2.2 Measurement procedure

The recoil energy of the fusion-evaporation reaction products delivered from SHIP was 35 – 40 MeV. To enable their efficient stopping and thermalization, a titanium foil with a thickness of $3.6 \mu\text{m}$ and a thickness variation of $\sim 0.4 \mu\text{m}$ was mounted at the entrance window and

the cryogenic gas cell was filled with 7 mbar of helium at 40 K. All reaction products were extracted mainly as doubly-charged ions and they were cooled and bunched inside the RFQ buncher at helium pressures around $5 \cdot 10^{-3}$ mbar. From there, the ions were transferred to the purification trap where the buffer-gas cooling technique described in Section 2.4.2 was employed to select the isotope of interest and prepare it for a cyclotron-frequency measurement in the measurement trap. A double pattern of the PI-ICR technique, which is described in Section 2.4.3, was used to project the ion motion on the position-sensitive detector. To calibrate the magnetic field, a reference ion, $^{133}\text{Cs}^+$, delivered from the surface ion source was used.

During the beamtime, a certain measurement sequence was followed. First, the magnetron and the reduced-cyclotron motion of $^{133}\text{Cs}^+$ were projected onto the detector. In the following measurement, the center of the measurement trap was projected using $^{133}\text{Cs}^+$. These measurements usually took 5 min each. Then, the magnetron and the reduced-cyclotron phase of the ion of interest were measured. Depending on the detection rate, this could take several hours and once it was completed, the measurement sequence would proceed with $^{133}\text{Cs}^+$ measurements. The magnetic field calculated from the cyclotron frequency of $^{133}\text{Cs}^+$ measured over the entire beamtime is shown in Fig. 4.1. During the calcium block, the ground states of $^{251,254}\text{No}$ and $^{254-256}\text{Lr}$ were measured, as well as their isomeric states (except for ^{256}Lr that does not have an isomer). A few days in the titanium block were used to measure ^{257}Rf . The sequence and the length of these measurements are displayed in Fig. 4.1.

4.2.3 Temperature and pressure stabilization

Due to low production rates of some isotopes, the measurement times were several hours long. The achieved precision in this case is strongly dependent on the accurate determination of the magnetic-field strength at the time when the nuclide of interest is measured (see Section 2.4.5). The magnetic-field strength is estimated from the reference measurements by employing interpolation methods described in Section 5.3

Figure 4.1: Magnetic field variation in the measurement trap determined from the evolution of the cyclotron frequency of the reference ion $^{133}\text{Cs}^+$ during the experimental campaign in 2018. The nominal value of the magnetic field is $B = 7.0540\text{ T}$ and the small variations are shown on the y axis. The periods dedicated for measurements of nobelium, lawrencium and rutherfordium isotopes are indicated by different colors.

which are sensitive to non-linear fluctuations of the magnetic field sustained by the superconducting solenoid. These changes of the magnetic field can be minimized by stabilizing environmental conditions (see Section 2.4.4). Therefore, the pressure and temperature stabilization systems were implemented at SHIPTRAP and the resulting magnetic-field fluctuations can be seen in Fig. 4.1.

Changes in the pressure inside the helium cryostat are correlated with the changes in the temperature of the liquid helium which leads to magnetic-field fluctuations. To reduce the systematic uncertainty introduced by these fluctuations, a pressure stabilization system is inte-

Figure 4.2: Fluctuations in the pressure inside the He cryostat (a), the temperature in the magnet bore (b), and the temperature of the power supplies (c) during the measurement time of ^{251}No . For details, see [text](#).

grated. A controller unit continuously compares the measured pressure with the set value and, in the case of deviations, the gas flow from the cryostat through a needle valve is adjusted (for details, see Ref. [77]). As an example, the pressure in the helium cryostat during the measurement period of ^{251}No is shown in Fig. 4.2 a. The pressure was stabilized to a mean value of 1017.59 mbar with a standard deviation of 0.07 mbar. The spikes are related to the liquid-helium level monitoring for which a resistance wire is used. This wire heats up during the measurement causing a small increase in pressure.

Without active temperature stabilization, the temperature in the magnet bore follows day/night ambient temperature fluctuations in the experimental hall, which are on the order of few $^{\circ}\text{C}$. Such deviations are similarly observed in the magnetic field intensity and, therefore, an in-house developed regulation system was implemented to stabilize the temperature [77]. Via a proportional–integral–derivative controller (PID controller), the temperature of the magnet bore is constantly monitored and compared with a set value that is chosen to be higher than the temperature of the experimental hall. If the deviation would exceed a certain threshold, the temperature would be corrected by adjusting correspondingly the power of a heating element connected to the air flow into the magnet bore. In Fig. 4.2 b the changes in the temperature of the magnet bore are shown during the measurement period of ^{251}No . The temperature was stabilized to a mean value of 36.15°C with a standard deviation of only $4\text{ m}^{\circ}\text{C}$. The two spikes visible in Fig. 4.2 b originate from an electronic disturbance in the temperature readout.

Similarly, a temperature stabilization is integrated for the switchable power supplies. These power supplies are used to set the trap potential and the stability of their output is crucial to ensure a harmonic electric field over time (Section 2.4.4). As shown in Fig. 4.2 c, during the measurement of ^{251}No , the temperature was stabilized at 44°C with a standard deviation of 0.03°C . The oscillations in the temperature of the switchable power supplies are the result of the day/night temperature fluctuations of the air in the experimental hall.

Data analysis

In this chapter, the analysis of the raw data obtained during the experimental campaign 2018 introduced in Section 4.2 is outlined. In this experiment, the PI-ICR technique was employed (see Section 2.4.3). The first step in the analysis is the background suppression, followed by the determination of the cyclotron frequency of an ion of interest and a reference ion. From these frequencies, mean cyclotron-frequency ratios are calculated using a linear interpolation and a polynomial method. Several sources of systematic uncertainties are investigated. The final results are presented in Chapter 6.

5.1 Background suppression

A typical PI-ICR image collected by the delay-line position-sensitive MCP detector at SHIPTRAP (for details see Section 3.6.3) is shown in Fig. 5.1. Every ion that is recorded by the detector is presented as a dot. In order to discriminate between signals produced by the ions of interest and noise signals, that may appear, for example due to dark counts of the detector or arise from contaminant ions, it is necessary to apply certain gates to the collected ion spectra.

Figure 5.1: From raw data to clean signal. Magnetron phase (upper images) and modified-cyclotron phase (lower images) after applying a ToF gate and a position gate. For all images, the number of ions per ejection was limited to one.

These gates are related to the number of detected ions, the ToF distribution and the position distribution of the detected ions. A LabView-software [78] is used to apply the cuts and thus filter out the data according to user-defined criteria. In this work, due to the low ion rates, the probability to simultaneously trap more than one ion is negligible. The measured ToF distributions of the ions ejected from the measurement trap were fitted using a Gaussian distribution. The ion events falling within 2σ width around the mean value are considered for further analysis. After applying the ToF gate, the obtained spectra were much cleaner allowing identification of the ion clusters. The ion clusters on the detector represent projections of the radial distribution of the ion cloud inside the trap which has the full width at a half maximum

of around $90\text{ }\mu\text{m}$ (see Section 2.4.2). Considering that the magnification factor was roughly 20, the full width at a half maximum of the detected ion clusters is estimated to be $\sigma \approx 2\text{ mm}$. The distribution of the ion cluster is described by a 2-dimensional Gaussian distribution. A cut in position with all the ion events falling within 2σ with respect to the distribution mean value is chosen. Figure 5.1 shows the magnetron and the modified-cyclotron phase projections after applying the gates in ToF and position.

5.2 Cyclotron frequency determination

As described in Section 2.4.3, the cyclotron frequency ν_c is given by

$$\nu_c = \frac{\phi + 2\pi(n_- + n_+)}{2\pi t}, \quad (5.1)$$

where n_- and n_+ are the numbers of full revolutions in the total phase accumulation time t . ϕ represents the difference between the magnetron and the modified cyclotron phase. The numbers of revolutions, which are integer values, and the phase accumulation time are known quantities as they are set by the user. The uncertainty of the latter one can be neglected since the time is set by frequency generators that are locked to the 10 MHz-signal of a rubidium clock with a frequency instability below $2 \cdot 10^{-11}$ over 1 s. However, it is important to note that due to the clock frequency of the function generator that is 250 MHz, the phase accumulation time can be set only in steps of 4 ns. The uncertainty of the cyclotron frequency $\delta\nu_c$ is determined by the uncertainty in the phase difference $\delta\phi$

$$\delta\nu_c = \frac{\delta\phi}{2\pi t}. \quad (5.2)$$

In order to obtain the phase difference, and thus the cyclotron frequency, it is necessary to determine the position of the center-image projection and the position of the ion projection after magnetron and modified cyclotron excitations. The ion images are projected along the x and y

Figure 5.2: Projections of the trap-center image and the ν_- - and ν_+ -phase images along the x and y axis. The absolute positions of the images are determined by applying a Gaussian fit to x and y projections which are shown as histograms on the right panel.

axis and a Gaussian fit is applied to obtain the absolute positions of the center-image and the phases of radial motion in the $x-y$ plane from the center of the Gaussians (see Fig. 5.2). The corresponding uncertainties in the positions are taken as the standard uncertainties of the center of the Gaussians resulting in

$$\text{abs. center spot} = (X_{\text{center}} \pm \delta X_{\text{center}}, Y_{\text{center}} \pm \delta Y_{\text{center}}), \quad (5.3)$$

$$\text{abs. magnetron spot} = (X_m \pm \delta X_m, Y_m \pm \delta Y_m), \quad (5.4)$$

$$\text{abs. mod. cyclotron spot} = (X_c \pm \delta X_c, Y_c \pm \delta Y_c). \quad (5.5)$$

Once the absolute positions are known, the relative positions are given by

$$\text{rel. magnetron spot} = (\bar{X}_m \pm \delta \bar{X}_m, \bar{Y}_m \pm \delta \bar{Y}_m), \quad (5.6)$$

$$\text{rel. mod. cyclotron spot} = (\bar{X}_c \pm \delta \bar{X}_c, \bar{Y}_c \pm \delta \bar{Y}_c), \quad (5.7)$$

where $\bar{X}_i = (X_i - X_{\text{center}})$, $\bar{Y}_i = (Y_i - Y_{\text{center}})$ and $\delta \bar{X}_i = \sqrt{\delta X_i^2 + \delta X_{\text{center}}^2}$, $\delta \bar{Y}_i = \sqrt{\delta Y_i^2 + \delta Y_{\text{center}}^2}$ for $i \in [m, c]$.

The absolute phase angle for magnetron and modified cyclotron projection can be calculated by employing the $\text{atan2}(x, y)$ function (from “2-argument arctangent”) that takes the relative positions of the spots as its arguments and returns the angle in radians

$$\phi_{\pm} = \begin{cases} \text{atan2}(\bar{X}_i, \bar{Y}_i) & \text{for } \text{atan2}(\bar{X}_i, \bar{Y}_i) \geq 0, \\ 2\pi - |\text{atan2}(\bar{X}_i, \bar{Y}_i)| & \text{for } \text{atan2}(\bar{X}_i, \bar{Y}_i) < 0. \end{cases} \quad (5.8)$$

Finally, the phase difference is given by the difference in the absolute phase angles of the magnetron and the modified-cyclotron phase

$$\phi = (\phi_- - \phi_+), \quad (5.9)$$

with an uncertainty

$$\delta\phi = \sqrt{(\delta\phi_-)^2 + (\delta\phi_+)^2}. \quad (5.10)$$

Independent of the employed measurement scheme (see the single and the double pattern, Section 2.4.3), the cyclotron frequency for every measurement file is obtained by employing Equations 5.1 and 5.9. In the case of single-pattern measurements, the modified cyclotron phase is recorded while the magnetron phase is taken from the measurement closest in time that is collected under the same conditions, such as excitations, phase accumulation time, etc.

5.3 Mean cyclotron-frequency ratio determination

According to Eq. 2.74, if the two cyclotron frequencies are measured at the same time, the magnetic-field ratio cancels out and the mass of the ion of interest is proportional to the cyclotron-frequency ratio $R = \nu_c^{\text{ref}}/\nu_c^{\text{ioi}}$. However, in the case of the SHIPTRAP spectrometer,

the cyclotron frequency of the ion of interest and the reference ion are measured alternately. Due to temporal fluctuations of the magnetic field it is necessary to interpolate the magnetic field strength from the time of a reference measurement to the time of the measurement of the ion of interest. To extract the frequency ratio, either a linear or polynomial interpolation method can be used.

5.3.1 Linear interpolation method

The linear interpolation method is based on the assumption that the magnetic field changes slowly and smoothly with time and can be approximated by a linear function. Suppose that the cyclotron frequency of an ion of interest ν_c^{ioi} is measured at the time t_n as shown in Fig. 5.3 a. If the cyclotron frequency of a reference ion ν_c^{ref} is measured before (t_{n-1}) and after (t_{n+1}) the ion of interest, then it can be linearly interpolated to the measurement time of the ion of interest t_n

$$\nu_c^{\text{ref}}(t_n) = \nu_c^{\text{ref}}(t_{n-1}) + \frac{t_n - t_{n-1}}{t_{n+1} - t_{n-1}} \left[\nu_c^{\text{ref}}(t_{n+1}) - \nu_c^{\text{ref}}(t_{n-1}) \right]. \quad (5.11)$$

A single frequency ratio at the time t_n is given by

$$R_i(t_n) = \frac{\nu_c^{\text{ref}}(t_n)}{\nu_c^{\text{ioi}}(t_n)}. \quad (5.12)$$

However, non-linear time-dependent variation of the magnetic field are not covered by a linear interpolation and if the deviations from the linear behavior are estimated (see Section 5.4.3) they can be considered in the overall uncertainty of a single cyclotron-frequency ratio δR_i given by

$$\delta R_i(t_n) = \sqrt{\left(\frac{\delta \nu_c^{\text{ref}}(t_n)}{\nu_c^{\text{ioi}}(t_n)} \right)^2 + \left(\frac{\nu_c^{\text{ref}}(t_n) \delta \nu_c^{\text{ioi}}(t_n)}{\nu_c^{\text{ioi}}(t_n)^2} \right)^2 + \xi_B^2 R_i(t_n)^2}, \quad (5.13)$$

Figure 5.3: Schematic diagram explaining the linear interpolation method (a) and the polynomial method (b) that are used for the determination of the cyclotron-frequency ratio. For details, see text.

where $\delta\nu_c^{\text{ref}}$ and $\delta\nu_c^{\text{ioi}}$ are the uncertainties attributed to the interpolated reference cyclotron frequency and the cyclotron frequency of the ion of interest, respectively, while ξ_B denotes the uncertainty that is attributed due to time-dependent non-linear magnetic field fluctuations. The mean cyclotron-frequency ratio R is given as the weighted mean of N single

cyclotron-frequency ratios R_i

$$R = \frac{\sum_N \omega_i R_i}{\sum_N \omega_i}, \quad (5.14)$$

where $\omega_i = \frac{1}{\delta R_i}$ is a weighting factor. The standard error of the weighted mean (internal error) and the biased weighted mean variance (external error) are given by

$$\delta R_{\text{int}} = \sqrt{\frac{1}{\sum_N \omega_i}}, \quad (5.15)$$

$$\delta R_{\text{ext}} = \sqrt{\frac{\sum_N \omega_i (R_i - R)^2}{\sum_N \omega_i (N - 1)}}. \quad (5.16)$$

The final uncertainty δR is calculated by adding a mass-difference-dependent uncertainty δ_M and a residual uncertainty ξ_{res} (see Sections 5.4.4 and 5.4.6) to the inner uncertainty

$$\delta R = \sqrt{\delta R_{\text{int}}^2 + \left(\Delta \frac{m}{q} \delta_M R\right)^2 + \left(\xi_{\text{res}} R\right)^2}. \quad (5.17)$$

5.3.2 Polynomial method

The polynomial method for the determination of the mean cyclotron-frequency ratio is schematically presented in Fig. 5.3 b. It is based on the approximation of changes in the cyclotron frequency by a polynomial of n -th order

$$\nu_c(t) \approx \sum_{i=0}^n a_i t^i. \quad (5.18)$$

The determination of the cyclotron-frequency ratio is performed by simultaneously fitting a polynomial function to both cyclotron-frequency sets, corresponding to the ion of interest $P_1(t)$ and the reference ion $P_2(t)$

$$P_1(t) = \sum_{i=0}^n a_i t^i, \quad (5.19)$$

$$P_2(t) = R \cdot P_1(t). \quad (5.20)$$

The maximum order of polynomial n depends on the number of data points N and it is given as $n = N - 1$. The proportionality between the two fits determines the cyclotron-frequency ratio R . The choice of polynomial order is based on the χ^2_{red} -criteria that is given by

$$\chi^2_{\text{red}} = \frac{1}{F} \sum_{i=0}^N \frac{\left(\nu_1(t_i) - P(t_i) \right)^2}{\delta \nu_1^2}, \quad (5.21)$$

where ν_1 denotes the cyclotron frequency with an uncertainty $\delta \nu_1$, P is a fitted polynomial function and F is the number of degrees of freedom. The best agreement between the fitted function and the data are achieved for $\chi^2_{\text{red}}=1$, which gives the best value for the cyclotron-frequency ratio $R = R_0$. The statistical uncertainty δR_{stat} is defined by

$$\chi^2_{\text{red}}(R_0 + \delta R_{\text{stat}}) - \chi^2_{\text{red}}(R_0) = 1. \quad (5.22)$$

A description of the polynomial method for determination of the mean cyclotron-frequency ratio can be found in Ref. [40], while Ref. [79] contains a fitting routine that was adopted for this analysis. As in the case of the linear interpolation method, aside from the statistical uncertainty δR_{stat} , the final uncertainty δR includes a mass-difference-dependent

uncertainty δ_M and the residual uncertainty ξ_{res} (see Sections 5.4.4 and 5.4.6)

$$\delta R = \sqrt{\delta R_{\text{stat}}^2 + \left(\Delta \frac{m}{q} \delta_M R\right)^2 + \left(\xi_{\text{res}} R\right)^2}. \quad (5.23)$$

5.4 Systematic uncertainties

5.4.1 Single-ion files

As discussed in Section 5.2, the phase spots are fitted using a Gaussian distribution. The extracted mean value corresponds to the center position of the spot and the uncertainty is given by the standard mean uncertainty u

$$u = \frac{\sigma}{\sqrt{n}}, \quad (5.24)$$

where σ denotes the standard deviation and n is the sample size. In the case of single-ion files, that are related to the measurements of the ion of interest where due to low rates only one ion is detected, it is not possible to fit a Gaussian distribution. Here, the coordinates are extracted from the ion position on the detector. Since for single-ion files $n = 1$, according to Eq. 5.24, the uncertainty is given by the standard deviation. In order to estimate the typical uncertainties (δX and δY), high-statistics phase images collected in online and offline measurements are evaluated and the mean of standard deviation from the Gaussian fits is taken as the uncertainty for single ion files. This corresponds to $\delta X = \delta Y = 1.2$ mm. The details of the evaluation can be found in Ref. [80].

5.4.2 Fluctuations of the center-image

The center-image on the detector corresponds to the center of the measurement trap. The center-images were recorded according to pattern

1 and pattern 2 presented in Fig. 2.13 without applying any excitation. This implies that the center-image position does not depend on a mass-over-charge ratio, m/q . Figure 5.4 confirms that there is no correlation between the position of the center-image and the mass-over-charge ratio. This is important since, due to low count rates of the ion of interest, the center-image projections are recorded by using the reference ion $^{133}\text{Cs}^+$. However, fluctuations in the position of the center-image are visible and present for the same ion species, which also concerns the $^{133}\text{Cs}^+$ center-image projections that are collected during the experiments. As an example, Fig. 5.5 shows the fluctuations in the X and Y positions of the center-image projection collected during the measurement block of $^{254}\text{Lr}^{2+}$. For this reason, the positions of the center-image (magenta and green data points) were linearly interpolated to the time of measurement of the ion of interest (black and red data points), as in the case of the cyclotron frequency of the reference ion. The uncertainty that is attributed to the position is taken as the standard deviation of coordinates measured in a certain block.

Figure 5.4: Coordinates of center-image projections in $x - y$ plane for different mass-over charge ratios.

Figure 5.5: Coordinates of center-image projections recorded using pattern 1 (x_1, y_1) and pattern 2 (x_2, y_2) without applying the RF excitations (for details, see Section 2.4.3). The centers were recorded during the measurement block of $^{254}\text{Lr}^{2+}$ and they are shown as magenta and green data points for x and y projections of $^{133}\text{Cs}^+$. The coordinates are linearly interpolated to the measurement time of the ion of interest and they are shown as black and red data points in both projections. The standard deviation of coordinates in each projection is assigned as position uncertainty.

5.4.3 Time-dependent non-linear magnetic field fluctuations

In the linear-interpolation method (see Section 5.3.1), the cyclotron frequency of the reference ion is interpolated to the measurement time of the ion of interest. However, the magnetic field can vary slightly from its value during the measurements although a stabilization is applied and its steadily linear variation is already taken into account. Therefore, a temporal non-linear component of the magnetic field drift has to be taken into account [49]. In order to estimate these time-dependent non-linear magnetic field fluctuations, a series of 30 min-long measurements of $^{133}\text{Cs}^+$ was performed over five days. The difference between the measured cyclotron frequency ν_c and the interpolated frequency ν_c^{lin} was used as a measure for the non-linearity of the magnetic-field drift $\Delta B = B - B_{\text{lin}}$ [49]. The relative standard deviation of the magnetic field as a function of the time ΔT elapsed between the two reference measurements is given by

$$\sigma((B - B_{\text{lin}})/B)(\Delta T) = \sigma((\nu_c - \nu_c^{\text{lin}})/\nu_c)(\Delta T) = \sqrt{\frac{\sum_N (R_i - R)^2}{N - 1}}, \quad (5.25)$$

where R_i is calculated as $(\nu_i - \nu_i^{\text{lin}})/\nu_i$ and R is the weighted mean of N cyclotron-frequency ratios. Figure 5.6 shows the change in the relative standard deviation of the magnetic field with increasing time ΔT between the two reference measurements.

The data were fitted with a linear function

$$\xi_B = \Delta T \cdot \delta_B, \quad (5.26)$$

from which a slope of $\delta_B = 1.4(3) \cdot 10^{-9} \text{ h}^{-1}$ was extracted. This value is added to the statistical uncertainty of each single cyclotron-frequency ratio according to Eq. 5.13.

Figure 5.6: Relative standard deviation of the magnetic field B with respect to the interpolated value B_{int} as a function of different time intervals ΔT between the two reference measurements of $^{133}\text{Cs}^+$.

5.4.4 Mass-difference dependent shift

The mass-difference dependent shift refers to the shift introduced through the measurement of the cyclotron frequency of two ions that have different mass-over-charge m/q ratios. If there are some imperfections related to the electric and/or magnetic field, then the two ions species will experience slightly different measurement conditions since they are probing slightly different volumes of the measurement trap.

In order to account for such a frequency shift, a set of cyclotron-frequency-ratio measurements, isotope 1 versus isotope 2, shown in Table 5.1 was evaluated. The ratios were compared to the literature values [81] and plotted as a function of the mass difference between two measured isotopes (see Fig. 5.7). The data were fitted using a linear function

$$\xi_M = \delta_M \Delta\left(\frac{m}{q}\right), \quad (5.27)$$

Table 5.1: A set of measurements performed before the beamtime to study mass-difference dependent shift.

Isotope 1	Isotope 2	$\Delta \frac{m}{q}$
$^{120}\text{Sn}^+$	$^{119}\text{Sn}^+$	1
$^{118}\text{Sn}^+$	$^{116}\text{Sn}^+$	2
$^{118}\text{Sn}^+$	$^{112}\text{Sn}^+$	6
$^{120}\text{Sn}^+$	$^{112}\text{Sn}^+$	8
$^{123}\text{Sb}^+$	$^{133}\text{Cs}^+$	10
$^{124}\text{Sn}^+$	$^{112}\text{Sn}^+$	12
$^{120}\text{Sn}^+$	$^{133}\text{Cs}^+$	13
$^{211}\text{Pb}^{2+}$	$^{133}\text{Cs}^+$	27.5

Figure 5.7: The difference of experimentally measured cyclotron-frequency ratios and ratios obtained from literature [81] as a function of the mass difference between isotope 1 and isotope 2 (see Table 5.1). The straight line is a linear fit to the data.

from which a slope of $\delta_M = 3.6(46) \cdot 10^{-10} (u/e)^{-1}$ was extracted. Here, u is the atomic mass unit and e is the elementary charge. This shift was included in final uncertainty using Eqs. 5.17 and 5.23.

5.4.5 Distortion of the ion-orbit projection

If the magnetron and the modified cyclotron motions are projected on the detector for different phase accumulation times t , starting from 0 to the period of the corresponding radial motion, then the ion-orbit image can be recorded. In Ref. [3] it is shown that an angular and/or a linear displacement between the symmetry axes of the magnetic field, the trap electrodes, and the detector can introduce a distortion of the ion-orbit projection and a shift of the center of the ion-orbit image with respect to the symmetry axis of the detector. As a consequence, this introduces a shift in the radial frequency ν due to the difference between the real angle ϕ , from an undistorted image, and the measured angle ϕ^* , from distorted image. The main impact on the frequency shift comes from the inclination of the detector symmetry axis with respect to the symmetry axis of the trap electrodes by an angle δ [3]. In this case, the ion-orbit image has an ellipsoidal shape and from the ratio of the major to the minor radius it is possible to calculate δ as $r_{\max}/r_{\min} = 1/\cos\delta$.

In order to estimate the value of the angle δ , all projections of the magnetron and the modified cyclotron motion recorded for $^{133}\text{Cs}^+$ during the beamtime 2018 were plotted and fitted by an ellipse (see Fig. 5.8). From the fits of the magnetron-phase projection (blue) and modified cyclotron-phase projection (orange), the inclination angles of 2.5° and 3.5° , respectively, were deduced. By including the inclination angles deduced from the fits into Eq. 2.71, for an arbitrary β , the relative shift in the cyclotron frequency is estimated to be well below $10^{-10} [\text{s}]/t$ and, hence can be neglected in the present analysis.

5.4.6 Residual uncertainty

To account for residual uncertainties arising from yet unknown effects that result in a systematic error, the data set used to estimate the time-dependent non-linear magnetic field fluctuations was evaluated. The cyclotron frequency of $^{133}\text{Cs}^+$ was measured in intervals of 30 min over a period of five days. This was the longest data set that was recorded without interruptions, with stable conditions concerning the tempera-

Figure 5.8: Projections of the magnetron (blue points) and the modified cyclotron (orange points) motions on the position-sensitive detector that are recorded during the whole beamtime. The blue and orange ellipses represent fits to the data from which the detector inclination with respect to the symmetry axis of the trap electrodes is deduced in order to estimate the shift in the frequency due to distortion of the ion-orbit image.

ture of the magnet bore, $\sigma(T_{\text{bore}}) = 0.03 \text{ K}$, and the pressure inside the liquid He vessel of the superconducting magnet, $\sigma(p_{\text{LHe}}) = 0.02 \text{ mbar}$. However, unlike during the beamtime, the temperature stabilization of switchable power supplies, that are used to set the trap potential, was not active and deviations within $\sigma(T_{\text{sw}}) = 0.87 \text{ K}$ were observed.

In Fig. 5.9, it is shown how the position of the center-image projection was affected by the changes in the ambient conditions. The temperature of the switchable power supplies and its fluctuation is directly connected to the stability of their output voltage and, thus, the stability of the electric field that defines the trap potential. A clear linear correlation between the position of the center-image and the temperature of the switches is visible, with Pearson's correlation coefficients higher than

Figure 5.9: A pair plot showing how the position of the center-image projection is influenced by changes in temperature of the magnet bore, temperature of the switches and pressure inside the liquid helium vessel. The panels on the diagonal are one-dimensional projections.

Figure 5.10: The heat maps containing Pearson's correlation coefficients that show the strength of correlation/anti-correlation between the ambient conditions and projected positions of center-image (a) and radial and angular distribution (b).

0.5, indicating a strong correlation (see Fig. 5.10 a). On the other hand, an anti-correlation was observed for the radial and angular position of the magnetron-phase projection with respect to the temperature of the switches (Fig. 5.10 b). The correlation studies can serve as a tool to check the impact of the ambient conditions on the measurement and to set requirements concerning stabilization parameters.

In order to quantify the residual uncertainty attributed to the measured cyclotron-frequency ratios ξ_{res} , the single cyclotron-frequency ratio of two adjacent $^{133}\text{Cs}^+$ frequencies was calculated. The obtained single cyclotron-frequency ratios from the whole data set are plotted in Fig. 5.11. By employing the linear interpolation method and considering the time-dependent non-linear magnetic field fluctuations, the mean cyclotron-frequency ratio was calculated. The deviation of the mean cyclotron-frequency ratio from 1 was taken as the residual uncertainty and it leads to $\xi_{\text{res}} = 1 - R = 1.2 \cdot 10^{-10}$. This value is overestimated when it comes to the conditions during the beamtime,

Figure 5.11: The resulting frequency ratios from $^{133}\text{Cs}^+$ versus $^{133}\text{Cs}^+$ measurements as a function of measurement time.

since the temperature stabilization of the switches was active. Nevertheless, the value is below the level of precision relevant for this work and thus, it can be neglected in Eq. 5.17 and Eq. 5.23.

5.4.7 Summary of systematic uncertainties

Table 5.2 presents an overview of the systematic uncertainties discussed in the previous sections. The major contribution to the final uncertainty comes from the time-dependent non-linear magnetic field fluctuations and the mass-difference dependent shift.

Table 5.2: Summary of systematic uncertainties.

Origin	Uncertainty
Non-linear magnetic field fluctuations	$1.4 \cdot 10^{-9} \text{ h}^{-1}$
Mass-difference dependent shift	$3.6 \cdot 10^{-10} / (u/e)$
Distortion of the ion-orbit projection	$10^{-10} [\text{s}] / t$
Residual uncertainty	$1.2 \cdot 10^{-10}$

Results and Discussion

During the experimental campaign described in Chapter 4, several isotopes of nobelium and lawrencium were measured at the SHIPTRAP mass spectrometer (see Chapter 3). Additionally, the first direct high-precision mass spectrometry of a superheavy element is performed on ^{257}Rf . The PI-ICR technique, introduced in Section 2.4.3, was employed for the first time in online experiments at SHIPTRAP under conditions of extremely low ion rates. Implementation of the PI-ICR technique and the improved performance of the cryogenic gas-stopping cell [51] enabled us to extend high-precision mass measurements in this region of the nuclear chart to more exotic isotopes and achieve a precision of $\delta m/m = 10^{-9}$. In addition to the measurements of the ground states, thanks to a mass resolving power of up to 11,000,000, it was possible to measure the excitation energies of low-lying isomers which were on the order of tens of keV. The results concerning investigated isomeric states can be found in Ref. [80].

This chapter focuses on the results for the ground-state masses of the investigated isotopes. The mean frequency ratios are given in Section 6.1. In Section 6.2, mass excesses from this work are compared to the data from the Atomic Mass Evaluation AME 2016 [81] and the

previous SHIPTRAP experiments performed in the region of the heaviest elements [5–7]. Finally, Section 6.3 shows how the presented results complement ongoing investigations on the nuclear structure around the deformed neutron shell gap at $N = 152$.

6.1 Mean frequency ratios of nobelium, lawrencium and rutherfordium isotopes

In this section, the mean cyclotron-frequency ratios of investigated isotopes and the reference ion are presented. Both approaches, a linear interpolation method and a polynomial method (introduced in Section 5.3) were used in the data analysis and the uncertainties are treated as described in Sections 5.3 and 5.4. The results obtained by using the two methods are compared with the results from Ref. [80] that considers the same data set. The final cyclotron-frequency ratio is given as a weighted mean of the values obtained in this work and the value in Ref. [80].

6.1.1 Nuclide ^{251}No

The first direct high-precision mass spectrometry of ^{251}No was performed within this work by employing the PI-ICR technique. The cyclotron frequency of $^{251}\text{No}^{2+}$ was measured with respect to the reference ion $^{133}\text{Cs}^+$. The first measurement was performed at a relatively short phase-evolution time of 20 ms in order to test the setup and measurement procedure. In order to resolve the isomeric state, with an excitation energy of about 105 keV, which corresponds to the cyclotron-frequency difference of ~ 0.4 Hz, the phase-evolution time was increased to 500 ms in the following measurements.

Figure 6.1 a shows the single cyclotron-frequency ratios of $^{133}\text{Cs}^+$ and $^{251}\text{No}^{2+}$ obtained by employing the linear interpolation method. In the same graph, the mean frequency ratio and the 1σ uncertainty band are represented by the solid orange line and the shaded area, respectively. A mean frequency ratio of $R = 0.944614690(8)$ is obtained from the

Figure 6.1: Temporal evolution of the single cyclotron-frequency ratios (a) and the cyclotron frequency measurements (b) of $^{133}\text{Cs}^+$ and $^{251}\text{No}^{2+}$. The first data point corresponds to a low-resolution and low-precision measurement at short phase-evolution time. (a) The linear interpolation method was applied to determine the single cyclotron-frequency ratios R_i . The solid line and the shaded band are the mean cyclotron-frequency ratio $R = 0.944614690(8)$ and its 1σ uncertainty, respectively. (b) A simultaneous polynomial fit represented by the dashed lines was applied to both data sets in order to extract the mean cyclotron-frequency ratio from the ratio of the applied polynomial functions leading to $R = 0.944614687(7)$.

linear interpolation method. Figure 6.1 b shows the measured cyclotron frequencies of $^{133}\text{Cs}^+$ and $^{251}\text{No}^{2+}$ as a function of time. A sixth-order polynomial function, presented by the green and the red dashed lines, is fitted to the measured cyclotron frequencies for both isotopes. The proportionality factor between the fit functions leads to a cyclotron-frequency ratio of $R = 0.944614687(7)$.

6.1.2 Nuclide ^{254}No

In this work the cyclotron frequency of $^{254}\text{No}^{2+}$ was measured for the first time using PI-ICR technique. The experimental campaign started with this measurement since ^{254}No was used to optimize the measurement procedure. This is due to the relatively high production cross section compared to the other isotopes that were investigated in this work. The measurement of $^{254}\text{No}^{2+}$ was repeated later during the experimental campaign with the aim to access the isomeric state with a half-life of about 250 ms (see Section 4.2). Both measurement blocks are considered in the analysis. The measurements were performed with respect to the reference ion $^{133}\text{Cs}^+$ with phase-evolution times ranging from 20 ms to 1200 ms.

Figure 6.2 shows the single frequency ratios of $^{133}\text{Cs}^+$ and $^{254}\text{No}^{2+}$ obtained in both measurement blocks. The data were analyzed by employing the linear-interpolation method. A mean cyclotron-frequency ratio was obtained as the weighted mean of all single frequency ratios. The solid orange line indicates the mean cyclotron-frequency ratio while the shaded area represents the 1σ uncertainty. The employed linear-interpolation method leads to a mean frequency ratio of $R = 0.955908553(6)$. The data analysis was performed also by using the polynomial method. Figure 6.3 shows the cyclotron frequencies of $^{133}\text{Cs}^+$ and $^{254}\text{No}^{2+}$ measured within two blocks. In the first block, a third-order polynomial function represented by dashed lines in Fig. 6.3 a was used to simultaneously fit the data points. The ratio between the two polynomial functions gives the mean cyclotron-frequency ratio. The same procedure is applied to the data collected in the second measurement block shown in (b). These data points are fitted with a first-order

Figure 6.2: Single cyclotron-frequency ratios of $^{133}\text{Cs}^+$ and $^{254}\text{No}^{2+}$ from the two measurement blocks obtained by employing the linear interpolation method. A mean cyclotron-frequency ratio is calculated to be $R = 0.955908553(6)$. Legend is the same as in Fig. 6.1 (a).

polynomial function. The weighted mean value is calculated from the two ratios, corresponding to the two measurement blocks, leading to the mean cyclotron-frequency ratio of $R = 0.955908555(3)$ which is in agreement with the result obtained by the linear-interpolation method.

6.1.3 Nuclide ^{254}Lr

Within this work, the atomic mass of ^{254}Lr was measured for the first time directly. The measurement was separated into two blocks (see Section 4.2). Different phase-evolution times, ranging from 50 ms to 600 ms were used. Due to the occurrence of an isomeric state, only measurements where the ground state has been resolved from the isomeric state were taken into account in the data analysis.

The cyclotron frequency of $^{254}\text{Lr}^{2+}$ was measured with respect to the reference ion $^{133}\text{Cs}^+$. The single cyclotron-frequency ratios obtained using the linear interpolation method around the weighted mean ratio

Figure 6.3: Temporal evolution of the cyclotron frequency during the first (a) and the second (b) measurement block of $^{133}\text{Cs}^+$ and $^{254}\text{No}^{2+}$. The mean cyclotron-frequency ratio based on the polynomial method was calculated as the weighted mean of ratios obtained from (a) and (b) resulting in $R = 0.955908555(3)$. Legend is the same as in Fig. 6.1 (b).

Figure 6.4: Single cyclotron-frequency ratios (a) and temporal evolution of the cyclotron frequency measurements (b) of $^{133}\text{Cs}^+$ and $^{254}\text{Lr}^{2+}$. (a) A mean cyclotron-frequency ratio of $R = 0.955928736(11)$ is obtained by employing the linear interpolation method. (b) A mean cyclotron-frequency ratio of $R = 0.955928726(27)$ is obtained by employing the polynomial method. Legend is the same as in Fig. 6.1.

are presented in Fig. 6.4 a. The solid orange line indicates the mean cyclotron-frequency ratio while the shaded area represents the 1σ uncertainty. A mean frequency ratio of $R = 0.955928736(11)$ is obtained. Only the first data point shown in Fig. 6.4 was taken during the first measurement block. The temporal evolution of the cyclotron frequencies corresponding to $^{133}\text{Cs}^+$ and $^{254}\text{Lr}^{2+}$ collected during the second measurement block is shown in Fig. 6.4 b. The cyclotron frequencies of $^{133}\text{Cs}^+$ and $^{254}\text{Lr}^{2+}$ were fitted with an eighth-order polynomial function presented by the dashed lines. The ratio between the two polynomial functions leads to a cyclotron-frequency ratio of $R = 0.955928726(27)$.

6.1.4 Nuclide ^{255}Lr

The cyclotron frequency of $^{255}\text{Lr}^{2+}$ was measured with respect to the reference ion $^{133}\text{Cs}^+$ with phase-evolution times of 100 ms, 1200 ms and 1500 ms.

The difference between the single cyclotron-frequency ratios obtained by using the linear interpolation method and the weighted mean ratio is shown in Fig. 6.5 a. The solid orange line represents the mean frequency ratio, while the shaded band indicates the 1σ uncertainty. The linear interpolation method gives a mean frequency ratio of $R = 0.959691645(7)$. In Fig. 6.5 b, a simultaneous polynomial method is applied to fit the cyclotron frequencies of $^{133}\text{Cs}^+$ and $^{255}\text{Lr}^{2+}$. The ratio between the two fourth-order polynomial functions shown by the dashed lines leads to the mean frequency ratio of $R = 0.959691639(4)$.

Figure 6.5: Temporal evolution of the single cyclotron-frequency ratios (a) and the cyclotron frequency measurements (b) of $^{133}\text{Cs}^+$ and $^{255}\text{Lr}^{2+}$. (a) A mean cyclotron-frequency ratio of $R = 0.959691645(7)$ is obtained by employing the linear interpolation method. (b) A mean cyclotron-frequency ratio of $R = 0.959691639(4)$ is obtained by employing the polynomial method. Legend is the same as in Fig. 6.1.

6.1.5 Nuclide ^{256}Lr

In this work, a cyclotron frequency of $^{256}\text{Lr}^{2+}$ was measured with respect to the reference ion $^{133}\text{Cs}^{+}$ with typical phase-evolution times around 100 ms, 250 ms and 1200 ms.

Figure 6.6 a shows the difference between the single frequency ratios obtained by using the linear interpolation method and the weighted mean frequency ratio which is presented by the solid orange line. The shaded band shows the 1σ uncertainty. The analysis leads to the mean frequency ratio of $R = 0.963461019(12)$. In Fig. 6.6 b, the cyclotron frequencies of $^{133}\text{Cs}^{+}$ and $^{256}\text{Lr}^{2+}$ are shown. The data points are fitted with a ninth-order polynomial function presented by the dashed lines. From the ratio of the two polynomials, a frequency ratio of $R = 0.963460998(23)$ is extracted.

6.1.6 Nuclide ^{257}Rf

In the SHIPTRAP experimental campaign 2018, the first direct mass measurement of a superheavy nuclide, namely ^{257}Rf , was performed. The experiment was running over approximately one week utilizing a ^{50}Ti -ion beam. The delivered primary beam intensity was about a factor of four lower than expected. Additionally, a reduction in efficiency of the setup towards the end of the beamtime was observed. As a result, the detection rate was only about one detected ^{257}Rf event in 10 hours, which is about one order of magnitude lower than estimated.

Compared to the measurements on nobelium and lawrencium isotopes, the phase images of ^{257}Rf showed an increased background level, probably due to the longer measurement time. For this reason, in addition to the gates that include the number of ions and ToF (see Section 5.1), a gate on the radial and angular distributions was applied. The radial distribution is determined by the applied amplitude of the dipolar excitation in the measurement trap, while the angular distribution is connected to the mass of the ion of interest, the phase-evolution time and the strength of the magnetic field. Thus, based on the radial distribution of the reference ion, the estimated mass of the ion of interest

Figure 6.6: Temporal evolution of the single cyclotron-frequency ratios (a) and the cyclotron frequency measurements (b) of $^{133}\text{Cs}^+$ and $^{256}\text{Lr}^{2+}$. (a) A mean cyclotron-frequency ratio of $R = 0.963461019(12)$ is obtained by employing the linear interpolation method. (b) A mean cyclotron-frequency ratio of $R = 0.963460998(23)$ is obtained by employing the polynomial method. Legend is the same as in Fig. 6.1.

and the cyclotron frequency of the reference ion, it was possible to estimate the region of interest where clusters of ^{257}Rf would appear. Two completely independent data analyses were performed (this work and the one presented in Ref. [80]) resulting in the selection of the same events. Namely, only a total of five events of $^{257}\text{Rf}^{2+}$ were recorded in two files. The measurements were performed with respect to the reference ion $^{133}\text{Cs}^+$ with a phase-evolution time of 50 ms, which was insufficient to resolve the isomeric state with excitation energy of about 70 keV [82].

The difference between the single frequency ratios obtained by applying the linear interpolation method and the weighted mean frequency ratio is shown in Fig. 6.7 a. The solid line indicates the mean frequency ratio which is $R = 0.96724017(52)$ and the shaded band represents the 1σ uncertainty. In Fig. 6.7 b, the temporal evolution of the cyclotron frequencies of $^{133}\text{Cs}^+$ and $^{257}\text{Rf}^{2+}$ is shown. The data points were fitted with a second-order polynomial function shown by the dashed lines. According to the polynomial method, a cyclotron-frequency ratio was extracted from the ratio of the two polynomial functions leading to $R = 0.96724017(67)$.

6.1.7 Summary of cyclotron-frequency ratios

An overview of the mean cyclotron-frequency ratios of doubly-charged nobelium, lawrencium and rutherfordium isotopes with respect to the reference ion $^{133}\text{Cs}^+$ is given in Fig. 6.8. The results obtained in this work by employing the linear interpolation-method and the polynomial method are presented by black and red data points, respectively. Additionally, the mean cyclotron-frequency ratios obtained in the independent data analysis of the same data set (see Ref. [80]) by using the linear-interpolation method are included. For the investigated isotopes, the results agree well with each other within their 1σ uncertainty. Therefore, the final cyclotron-frequency ratios for each isotope were calculated as the weighted mean of the three results. The final results for the mean cyclotron-frequency ratio are given in Tab. 6.1. Figure 6.8 shows the difference between the frequency ratio values of nobelium,

Figure 6.7: Temporal evolution of the single cyclotron-frequency ratios (a) and the cyclotron frequency measurements (b) of $^{133}\text{Cs}^+$ and $^{257}\text{Rf}^{2+}$. (a) A mean cyclotron-frequency ratio of $R = 0.96724017(52)$ is obtained by employing the linear interpolation method. (b) A mean cyclotron-frequency ratio of $R = 0.96724017(67)$ is obtained by employing the polynomial method. Legend is the same as in Fig. 6.1.

lawrencium and rutherfordium isotopes obtained in this work and Ref. [80] and the final frequency ratios represented by the blue shaded area indicating the 1σ uncertainty. The statistical uncertainty was calculated as the average uncertainty of the value derived from the polynomial method and the two values from the linear-interpolation method. The systematic uncertainty originating from the mass-difference-dependent shift (see Section 5.4.4) is quadratically added to the statistical uncertainty.

Figure 6.8: The mean cyclotron-frequency ratios of doubly-charged nobelium, lawrencium and rutherfordium isotopes with respect to the reference ion $^{133}\text{Cs}^+$. The results obtained in this work (linear and polynomial method) and the results reported in Ref. [80] show good agreement within 1σ uncertainty. The final values of the mean cyclotron-frequency ratios are given as the weighted mean whose uncertainty is displayed by the shaded area.

Table 6.1: The mean cyclotron-frequency ratios of the investigated isotopes measured as doubly-charged ions with respect to the singly-charged ^{133}Cs . The corresponding mass excess values are calculated and compared to the values from the AME16 [81].

Isotope	R	ME [keV]	ME _{AME} [keV]
$^{251}\text{No}^{2+}$	0.9446146872(85)	82851.4(21)	82849(114)
$^{254}\text{No}^{2+}$	0.9559085534(53)	84733.4(13)	84723.3(97)
$^{254}\text{Lr}^{2+}$	0.955928732(16)	89729.5(41)	89871(301)
$^{255}\text{Lr}^{2+}$	0.9596916425(63)	89933.2(16)	89947(18)
$^{256}\text{Lr}^{2+}$	0.963461016(15)	91737.0(38)	91747(83)
$^{257}\text{Rf}^{2+}$	0.96724015(56)	95923(141)	95867(11)

6.2 Mass excess of nobelium, lawrencium and rutherfordium isotopes

According to Eq. 2.76, once the mean cyclotron-frequency ratio R is determined, it is possible to obtain the absolute mass of the atom m . Usually, the atomic masses are given in terms of mass excess ME that is calculated as $ME = [m - A \cdot u]c^2$, where A is the atomic mass number, c is the speed of light and u represents the atomic mass unit. The atomic mass unit is the mass of one twelfth of the mass of ^{12}C . Therefore, the mass excess of an atom shows its binding energy per nucleon with respect to the binding energy per nucleon of ^{12}C .

The mean cyclotron-frequency ratios of the investigated nuclei are summarized in Tab. 6.1. Based on these values the corresponding mass excess values are calculated and given together with the mass excess values from the Atomic Mass Evaluation AME 2016 [81]. Figure 6.9 shows the difference between the mass excess values of nobelium, lawrencium and rutherfordium isotopes of the atomic mass evaluation 2016 (black data points) and the results obtained in this work including their 1σ mass excess uncertainty (blue band). In addition, the results obtained in the

Figure 6.9: Mass excess difference between the masses taken from the AME16 [81] and the previous SHIPTRAP values [5–7], and the new SHIPTRAP results obtained in this work. The latter are displayed by the 1σ uncertainty shown by the blue shaded area.

previous SHIPTRAP experiments [5–7] using the Time-Of-Flight Ion-Cyclotron-Resonance (ToF-ICR) technique are shown for comparison (red and blue data points). These results have already been included in the AME16.

So far, the atomic mass of ^{251}No was known with an uncertainty of 114 keV [81]. Since ^{251}No decays through emission of an α particle to ^{247}Fm , its atomic mass has been derived from the Q_α value and the atomic mass of ^{247}Fm , which dominates the mass uncertainty of ^{251}No . The mass excess obtained in this work $ME = 82851.4(21)$ keV is in agreement with the AME value of $ME_{\text{AME}} = 82849(114)$ keV. Thus, the first direct Penning trap mass measurement of ^{251}No , performed in this work, has improved its mass uncertainty by a factor of 54. Additionally, the excitation energy of the isomeric state of $^{251\text{m}}\text{No}$ with a half-life of about 1 s was determined directly for the first time leading to the value of 105(3) keV [80]. This value agrees well with the indirectly obtained excitation energy of 106(7) keV which is derived from the estimated excitation energy of $^{247\text{m}}\text{Fm}$ and supports the decay scheme given in Ref. [83].

Due to its relatively high cross section, the isotope ^{254}No has been measured directly with high precision in the previous SHIPTRAP experiments [5–7] as well as at RIKEN using Multi-Reflection Time-Of-Flight Mass Spectrometry (MR-ToF MS) [84]. The mass excess value given by the atomic mass evaluation is $ME_{\text{AME}} = 84723.3(97)$ keV. As shown in Fig. 6.9, the mass excess $ME = 84733.4(13)$ keV obtained in this work is in agreement with the values reported in the previous SHIPTRAP experiments as well as with the atomic mass evaluation. Besides, the uncertainty in this work was reduced by one order of magnitude.

In the case of ^{254}Lr , the AME value is based on the atomic mass of its daughter, namely ^{250}Md , and the Q_α value. The relatively high mass uncertainty, 301 keV, is dominated by the mass uncertainty of ^{250}Md . In this work, the atomic mass of ^{254}Lr has been measured directly for the first time improving its precision by two orders of magnitude. The result obtained in this work $ME = 89729.5(41)$ keV is in good agreement with the AME mass excess value of $ME_{\text{AME}} = 89871(301)$ keV. Furthermore, the excitation energy of $^{254\text{m}}\text{Lr}$ was measured directly for the first time leading to the value of 110(6) keV [80]. The obtained result confirms existence of the proposed isomeric state [85] and it is in agreement with the indirectly derived excitation energy of 108(25) keV [86].

The mass of ^{255}Lr has already been measured directly at SHIPTRAP [6]. While in this experiment the aim was to take advantage of the high mass resolving power, which was achieved thanks to the PI-ICR technique, to precisely measure a low-lying isomeric state with an estimated excitation energy of about 37 keV [75], the ground state is measured in parallel with high precision. The direct determination of excitation energy leads to the value of 32(2) keV [80] which supports the decay scheme presented in Ref. [75]. Furthermore, the obtained mass excess is $ME = 89933.2(16)$ keV and it agrees with the AME value $ME_{\text{AME}} = 89947(17)$ keV. However, as shown in Fig. 6.9, the result disagrees with the previous SHIPTRAP value ($ME = 89958(16)$ keV) reported in Ref. [6]. Since this value is between the values obtained for the ground and the isomeric state in this work, a contribution from the isomeric state in Ref. [6] is a possible explanation for this deviation. Namely, because of the insufficient resolving power of the ToF-ICR technique, the resonance curves for the ground and isomeric states could not be resolved, which leaves the possibility for an isomeric contamination.

The first direct mass spectrometry of ^{256}Lr has already been performed at SHIPTRAP [6]. Due to the low production cross section, it was the most challenging measurement in the region of the heaviest elements performed using the ToF-ICR technique. A short excitation time of 200 ms and limited statistics resulted in a mass uncertainty of 83 keV. Thus, the atomic mass of ^{256}Lr was measured again in order to reduce mass uncertainty. The obtained mass excess of $ME = 91737.0(38)$ keV is in good agreement with the value of the AME16 $ME_{\text{AME}} = 91747(83)$ keV. In this work, the precision has been improved by a factor of 40. An isomeric state has not been observed in this experiment.

Finally, the first high-precision mass spectrometry of superheavy element was performed within this work. The isotope ^{257}Rf has been measured directly. Due to limited beamtime and a low efficiency, only a total of five events were recorded. Nevertheless, these events sufficed to determine the mass of the ground state of ^{257}Rf . The measurement was performed with a short phase-evolution time of 50 ms, which was

insufficient to resolve an isomeric state with an excitation energy of 70(15) keV [87]. From the obtained cyclotron-frequency ratio, the mass excess of $ME = 95958(139)$ keV was calculated. However, since an isomeric contamination cannot be excluded, for the final mass excess and its uncertainty the mixture of the two states has to be taken into account. The procedure described in AME16 [81] has been applied according to which the mass excess of the ground state is calculated as $ME = ME_{\text{exp}} - E_1/2$, where ME_{exp} and E_1 represent the measured mass and the excitation energy, respectively. The mass uncertainty is given by $\sigma^2 = \sigma_{\text{exp}}^2 + (\sigma_1/2)^2 + (0.29E_1)^2$ where σ_{exp} and σ_1 are uncertainties of the measured mass excess and the excitation energy, respectively. Therefore, our final result for the mass excess of ^{257}Rf is $ME = 95923(141)$ keV and, as shown in Fig. 6.9, it agrees with the AME mass excess value $ME_{\text{AME}} = 95867(11)$ keV.

6.3 Comparison of experimental results and theoretical models

Experimentally obtained mass excesses and their derivatives are quantities that are used to test theoretical nuclear mass models. Figure 6.10 shows the mass excesses of nobelium (a) and lawrencium (b) isotopes for neutron numbers in the range of $N = 146 - 160$. The black data points are taken from the AME16 [81], while the data obtained in this work are shown in red. To compare the experimental results with the theoretical predictions, the mass excess deduced from several theoretical models is plotted.

The models developed by Möller *et al.* [88] and Muntian *et al.* [89] are based on the microscopic-macroscopic approach which considers the liquid-drop model and include modifications by a microscopic shell-correction energy. The model of Muntian *et al.*, is optimized for heavy nuclei in the range of $Z = 102 - 109$, while the model from Möller *et al.* is a global finite-range droplet model. Both models reproduce well the trend of mass excesses for different isotopes of nobelium and lawrencium, although the agreement is better in the case of the microscopic-

Figure 6.10: The mass excess of nobelium (a) and lawrencium (b) isotopes. The black data points are taken from the atomic mass evaluation [81], while the red data points are from the present work. Theoretical predictions from [88–91] are presented by the dashed lines. For further details see text.

macroscopic model that is optimized for very heavy elements [89]. Figure 6.11 presents deviations between the mass excess values from this work and the AME16 and predictions based on the models developed

by Muntian *et al.* and Möller *et al.*. The predictions of the latter deviate from the AME16 and this work by several 100 keVs. According to the microscopic-macroscopic models, the island of stability is located at $Z = 114$, $N = 184$.

Figure 6.11: Mass-excess difference between the predicted values [88, 89] (orange and green data points) and the results of the AME16 [81] and this work (blue and pink 1σ uncertainty bands, respectively) for nobelium (a) and lawrencium (b) isotopes. For further details, see text.

The SkM* [90] and TW99 [91] curves in Fig. 6.10, show models that are examples of microscopic approaches in which the interactions between the single particles are described by a mean-field. Self-consistent mean-field models use energy density functionals that are based on effective nucleon-nucleon interactions. Here, the models using the Skyrme-Hartree-Fock SkM* [90] and TW-99 [91] effective interactions are shown. For the latter one, a relativistic mean-field model is used and the data are available only for even-even nuclei. The mass excess predicted by these models deviates from the experimental results by several MeV. The SKM* and the TW-99 models predict the center of the island of stability to be at $Z = 126$, $N = 184$ and at $Z = 120$, $N = 172$, respectively.

To investigate the nuclear shell structure, it is useful to look into quantities that can be derived from the mass excess. Such a quantity is, for example, the two-neutron separation energy S_{2n} , which is defined as the energy necessary to remove two neutrons from the nucleus and can be calculated as

$$S_{2n}(N, Z) = ME((N - 2), Z) + 2m_n c^2 - ME(N, Z), \quad (6.1)$$

where m_n is the neutron mass. A sudden change of the two-neutron separation energies at a specific neutron number may indicate a nuclear shell gap. This is visible in Fig. 6.12 where the two-neutron separation energies of nobelium (a) and lawrencium (b) isotopes are plotted. The black data are obtained from the AME16 [81], while the red data points indicate the contribution from this work. The predictions of the above-mentioned theoretical models [88–91] are included for comparison. The experimental data show a kink at neutron number $N = 152$ that is well reproduced by the microscopic-macroscopic models [88,89] and the self-consistent mean-field model SkM* [90]. This kink indicates the deformed neutron shell closure at neutron number $N = 152$ which was reported in Ref. [6] thanks to the precise mass measurements performed at SHIPTRAP.

Figure 6.12: The two-neutron separation energies of nobelium (a) and lawrencium (b) isotopes. The black data points are obtained from the atomic mass evaluation [81], while the red data points show the two-neutron separation energies where this work contributes by providing one or two mass values. Theoretical predictions from [88–91] are presented by the dashed lines. For further details see text.

A more sensitive indicator of shell closures is the two-neutron shell gap δ_{2n}

$$\begin{aligned}\delta_{2n}(N, Z) &= S_{2n}(N, Z) - S_{2n}(N + 2, Z) \\ &= ME((N - 2), Z) - 2ME(N, Z) + ME(N + 2, Z),\end{aligned}\tag{6.2}$$

which is used in Ref. [6] to map the nuclear shell effects around $N = 152$. Figure 6.13 shows an updated comparison between experimentally obtained and predicted δ_{2n} values for nobelium (a) and lawrencium (b) isotopes around $N = 152$. The red data points indicate the δ_{2n} values where this work contributes by providing at least one mass excess value used in the calculation. The trend of the shell-gap parameter is well reproduced by the microscopic-macroscopic models [88,89] and the self-consistent mean-field model SkM* [90] indicating a peak at the deformed $N = 152$ shell. The model based on the TW99 parameterization predicts deformed shell closure at $N = 154$.

Although there are strong deviations of the predicted mass excess, in general, the agreement in the strength of the two-neutron shell gap is significantly better. The microscopic-macroscopic model optimized for very heavy elements [89] shows the best agreement with the experimental results for the two-neutron shell gap, which is also in agreement with the results reported in Ref. [6]. In this work, the obtained neutron shell gaps for $N = 152$ are 1226.73(1) keV for nobelium and 1374(207) keV for lawrencium. The obtained values are few times lower compared to lighter nuclei such as doubly-magic ^{132}Sn ($Z = 50, N = 82$) and ^{208}Pb ($Z = 82, N = 126$) which indicates weaker shell closure in the region of the heaviest elements.

Figure 6.13: The two-neutron shell gap of nobelium (a) and lawrencium (b) isotopes. The black data points are obtained from the atomic mass evaluation [81], while the red data points show the two-neutron shell gap where this work contributes by providing at least one mass value. Theoretical predictions from [88–91] are presented by the dashed lines. For further details see text.

Another important quantity for nuclear models is the pairing energy. The pairing energy explains the increased stability for even-even nuclei due to formation of proton-proton and neutron-neutron pairs. For example, the neutron pairing energy Δ_{2n} can be calculated from the masses of three consecutive nuclides

$$\Delta_{2n}(N, Z) = (-1/2)^N [ME(Z, N - 1) - 2ME(N, Z) + ME(Z, N + 1)], \quad (6.3)$$

and it shows sharp peaks at certain values of neutron numbers indicating enhanced stability there. The neutron pairing energy of nobelium and lawrencium isotopes around $N = 152$ is plotted in Fig. 6.14. The red data points show the values that are influenced by this work, at least by one mass excess value, while the AME16 values are shown in black. Although all models predict the strongest pairing at the neutron number $N = 152$, the global model by Möller *et al.* [88] shows the best agreement with our results, followed by Muntian *et al.* [89]. The model utilizing the TW-99 effective interaction is not plotted since only data for even-even nuclei are available. The general trend of the neutron pairing energy as a function of nucleon number is the same for nobelium and lawrencium isotopes. The results obtained in this work show the maximum neutron pairing energy of 850(16) keV for nobelium and 800(3) keV for lawrencium reflecting the presence of a shell gap at $N = 152$. The latter value comes from the three measured masses of consecutive nuclides that are obtained in this work (mass measurements of ^{254}Lr , ^{155}Lr and ^{256}Lr). The neutron pairing energy in the region of doubly-magic ^{100}Sn ($Z = 50, N = 50$), ^{132}Sn ($Z = 50, N = 82$) and ^{208}Pb ($Z = 82, N = 126$) is on the order 2 – 4 MeV which is few times higher than in the case of nobelium and lawrencium isotopes indicating a higher level density in this region.

The experimentally obtained parameters presented above serve as an input for theoretical models aiming at mapping the shell effects in the region of the heavy and superheavy elements. To reproduce the measured masses and converge to common predictions concerning the shell closures, the parameters used in theoretical models have to be adjusted

Figure 6.14: The neutron pairing energy of nobelium (a) and lawrencium (b) isotopes. The black data points are obtained from the atomic mass evaluation [81], while the red data points show the neutron pairing energy where this work contributes by providing at least one mass-excess value. Theoretical predictions from [88–90] are presented by the dashed lines. For further details see text.

by taking into account experimental values. Direct high-precision mass measurements provide absolute masses and model-independent differential quantities that can reveal the structure of nuclei. Besides, the atomic masses of heavier nuclei can be derived indirectly by pinning down members of α -decay chains. The results obtained within this work for ^{251}No and ^{254}Lr provided the new anchor points in the region of heavy elements. These anchor points are important since in odd- A and odd-odd nuclei the decay from the mother nuclide usually populates an excited state of the daughter nuclide. Here, the excitation energies are not known precisely resulting in high mass uncertainty. As shown in Fig. 6.15, the new anchor points for odd- A isotope ^{251}No and odd-odd isotope ^{254}Lr in combination with results from decay spectroscopy will contribute to improving atomic masses of isotopes up to the element darmstadtium. Some of the members of these decay chains are too short-lived for direct high-precision PTMS or even for MR-TOF (for example ^{267}Ds).

Figure 6.15: Indirect mass determination of superheavy nuclei based on directly measured masses of ^{251}No and ^{254}Lr and α -decay energies.

Towards high-precision mass measurements of ^{100}Sn

Neutron-deficient medium-heavy nuclei near the $N = Z$ line are of great interest for nuclear and astrophysical studies. The proximity to the proton drip-line makes this region suitable for a better understanding of nuclear shell structure far from stability. The investigation of the astrophysical rapid proton-capture (rp) process which follows the $N = Z$ line requires input data coming from observed properties of these nuclei.

The Penning trap mass spectrometer SHIPTRAP (see Chapter 3) has delivered valuable results for neutron-deficient isotopes of medium-heavy to transuranium elements [6, 92, 93]. The relative precision that was achieved in these experiments is in the range of a few times 10^{-8} . Thanks to a number of technical and methodological improvements, the recent experimental campaigns dedicated to the region of heavy and superheavy elements demonstrated that despite the very low production rates, a high precision in mass determination is still achievable, as shown in Chapter 6. Consequently, one of the goals is to continue the successful program of mass measurements of neutron-deficient medium-

heavy nuclei near the $N = Z$ line that would contribute to nuclear astrophysics studies [92].

This chapter is dedicated to the motivation for high-precision mass measurements in the region of ^{100}Sn , the introduction of the main challenges that concern production and separation of ^{100}Sn , as well as the presentation of a possible solution that can be employed.

7.1 ^{100}Sn and its neighbours

In the chart of nuclides, ^{100}Sn is the heaviest nucleus with equal numbers of protons Z and neutrons N , making it a self-conjugate nucleus (see Fig. 7.1). Since it has a filled proton shell $Z = 50$ and a filled neutron shell $N = 50$, it is called a doubly-magic nucleus. According to the nuclear shell model [70, 71], the shells $g_{9/2}$ are completely occupied by protons and neutrons which leads to an increase in the nucleon pairing energy [8]. This system is well suitable to study the interaction between protons and neutrons occupying the same orbits [8]. For even-even nuclei, the average p-n interaction of the last nucleons δV_{pn} can be expressed by [94]

$$\delta V_{\text{pn}}(Z, N) = \frac{1}{4}[\{B(Z, N) - B(Z, N-2)\} - \{B(Z-2, N) - B(Z-2, N-2)\}], \quad (7.1)$$

where B stands for the binding energy of the nucleus. δV_{pn} for even-even nuclei refers to the interaction of the $(Z - 1)$ and Z^{th} protons with the $(N - 1)$ and N^{th} neutrons and is very sensitive to changes in shell structure in exotic nuclei. The double difference of binding energies embodied in δV_{pn} isolates the interaction of the last proton with the last neutron, subtracting off mean field and like-nucleon interactions [94].

From the binding energies, the two-proton separation energy and the two-neutron separation energy as well as the empirical shell gap can be derived. The first two quantities reveal nuclear shell closures and shape

Figure 7.1: Mass uncertainties of isotopes along the $N = Z$ line in the vicinity of ^{100}Sn from AME2016 [81] and the predicted path of the rp-process [95].

transitions while the shell gap (the energy difference between shells) indicates the strength of the shell closure. The separation energies are on the order of a few MeV while the shell gap is several 100 keV. To elucidate the structure of exotic nuclides based on the changes in the patterns of these quantities, the mass uncertainties of the involved nuclides should be less than several 10 keV. This corresponds to a relative precision of $\delta m/m = 10^{-7}$ for $A = 100$. In the region of ^{100}Sn the atomic masses of the nuclei closest to the drip line are not known with high precision. A part of the nuclear chart along the $N = Z$ line in which the mass uncertainties from the Atomic Mass Evaluation (AME) [81]

Figure 7.2: Schematic of the Gamow-Teller transition. The spins of the emitted positron and neutrino couple to total spin $S = 1$, while the change of the angular momentum of the nucleus is $\Delta J = 0, \pm 1$.

are indicated, is shown in Fig. 7.1. The mass of ^{100}Sn is known with a precision of 10^{-5} [81].

Another property that makes ^{100}Sn interesting is its β^+ decay. In this process, the doubly magic ^{100}Sn with spin 0^+ decays into the 1^+ state of ^{100}In [96]. During the decay, a positron-neutrino pair with parallel spins is emitted leading to total spin $S = 1$. Thus, the change in angular momentum from the initial into the final state is $\Delta J = 1$ (see Fig. 7.2). This type of decay is known as Gamow-Teller (GT) transition and it is characterized by a comparative half-life or $\log(ft)$ value. f represents the Fermi integral function which depends on the daughter atomic number Z' and the β -decay endpoint energy E_0 (the maximum positron energy), while t stands for the half-life of the transition [9]. An interesting fact about ^{100}Sn is that it has an extremely small $\log(ft)$ value. Once the ft value is known, it is possible to calculate the strength of the Gamow-Teller transition B_{GT} for ^{100}Sn as

$$B_{\text{GT}} = \frac{D}{ft}, \quad (7.2)$$

where D represents the axial-vector coupling constant that comes from the decay of a free neutron. Several experiments have been performed with the aim to extract the GT transition strength of ^{100}Sn by measuring the β -decay half-life and the endpoint energy. The experimental B_{GT} values obtained up to now are: $9.1^{+2.6}_{-3.0}$ [96], $5.8^{+5.5}_{-3.2}$ [97] and

$4.4^{+0.9}_{-0.7}$ [98]. While the last value is the most precise due to the improved statistics, it deviates from the value obtained in Ref. [96]. The difference comes mostly from different values of the endpoint energy that are: 3.29 ± 0.20 MeV [96], $3.8^{+0.7}_{-0.3}$ MeV [97] and 3.91 ± 0.15 MeV [98]. The large uncertainties for the B_{GT} value are dominated by the uncertainty in the value for the β endpoint energy. To improve the precision of the B_{GT} value it is necessary to measure the decay energy (Q -value) with an uncertainty below 100 keV as it enters the B_{GT} value approximately as Q^{-5} .

Additionally, $\log(ft)$ is inversely proportional to the square of the nuclear matrix element [9]. The nuclear wave functions before and after the decay enter the nuclear matrix element. Thus, Gamow-Teller transitions can be used as a tool to study the nuclear structure. The comparison of experimental and predicted GT-transition strengths in ^{100}Sn in Ref. [98] shows that most of the predicted values overestimate the experimental GT strength obtained in the same work. Due to the high uncertainties, the discrimination between different model calculations and the experimental values from Ref. [96] and Ref. [97] was not possible. Therefore, the precision of the B_{GT} value should be improved in order to test, guide, and constrain theoretical approaches. The β -decay energy has to be cross-checked and measured with higher precision. A possible solution is to determine the difference in masses of ^{100}Sn and ^{100}In .

From the perspective of nuclear astrophysics, the ^{100}Sn region is crucial for the predictions of the path of the rapid proton-capture process (rp-process). The rp-process is responsible for the creation of elements in the thermonuclear explosion at the surface of a neutron star accreting hydrogen-rich matter from a companion star and it is the source of so-called X-ray bursts [95]. As the name suggests, the rp-process is a sequence of rapid proton-capture reactions and subsequent slow β^+ decays. It starts from He and creates nuclides close to the $N = Z$ line. In a hydrogen-rich environment it can reach up to ^{107}Te and ^{108}Te , but due to α -instability, the process does not progress towards higher atomic numbers. Instead, the reaction flow returns to Sn isotopes creating the

Sn-Sb-Te cycle, which sets the endpoint of the rp-process [99]. The path of the rp-process in the region of ^{100}Sn is indicated in Fig. 7.1.

Proton separation energies are one of the most crucial parameters for the predictions of the path of the rp-process [92,100]. The proton separation energy can be derived from the mass difference of two nuclei differing by one proton. By now, more than 2/3 of all nuclides relevant for the rp-process have been measured with high precision [101]. However, one of the current problems for theoretical studies of the rp-process is that the mass values in the region of ^{100}Sn have high uncertainties due to the low production rates and difficulties in separation of these species at the existing radioactive beam facilities. The mass uncertainties should be reduced to a level of better than 10 keV to be able to make any progress in the exact prediction of the path of the rp-process [92,100].

A high-precision mass measurement of ^{100}Sn and its neighboring nuclei is of great interest not only to guide and improve nuclear models but also to understand the Gamow-Teller transition as well as for an exact prediction of the rp-process path. So far, the mass of ^{100}Sn has been only measured using the cyclotron technique at GANIL. The CSS2 cyclotron of GANIL was used as a high-precision spectrometer to determine the ToF of ^{100}Sn with respect to the reference ion ^{100}Ag [102]. In the region of light isotopes, this technique provides a mass resolution of less than 10^{-6} . However, the mass of ^{100}Sn was measured with a precision of 10^{-5} which was limited by low statistics (only 12 detected events) [102].

The required precision of $\delta m/m = 10^{-7}$ can be easily reached with the Penning trap mass spectrometry of ^{100}Sn . It is the most precise technique for mass measurements and it does not require a large number of detected ions. The most neutron-deficient tin isotopes that were measured using Penning traps are ^{104}Sn [92] and ^{103}Sn [103]. The experiments were conducted at the SHIPTRAP facility and the JYFLTRAP facility, respectively.

7.2 Production of ^{100}Sn

Experimental studies of ^{100}Sn are challenging due to the low production yield and the huge number of unwanted by-products. In the past, two experimental approaches were used for the production of ^{100}Sn : In-Flight fragmentation (IF) and fusion-evaporation reactions.

In the first method (IF), a heavy-ion primary beam with an energy 100 MeV/u - 1 GeV/u is impinged on onto a relatively thick target (typically a few g/cm²) of a light element such as beryllium. The reactions that can occur in this case are projectile fragmentation and Coulomb-induced fission. The reaction products are separated by utilizing magnetic and electric fields. The production cross section of the isotope ^{100}Sn using IF fragmentation in the experiments performed at GSI [96] and RIKEN [104] are presented in Tab. 7.1.

Another method is the production of radioactive isotopes in a fusion-evaporation reaction where a primary beam with energy of about 5 MeV/u impinges on a thin target (~ 0.5 mg/cm²) forming radioactive isotopes. Using this technique, the isotope ^{100}Sn has already been produced at GANIL [102]. As shown in Tab. 7.1, the production cross section of ^{100}Sn is three orders of magnitude higher than the one of the fragmentation reactions. However, the lower cross section in fragmentation reactions compared to fusion-evaporation reaction can be counter-balanced by thicker production targets.

The production yields of ^{100}Sn via fusion-evaporation reactions were investigated in Refs. [102] and [105]. According to these studies, the highest cross sections for production of ^{100}Sn have been observed in the following reactions: $^{58}\text{Ni}(^{46}\text{Ti}, 4n)^{100}\text{Sn}$ ($\sigma = 30$ nb, predicted value) [105] and $^{58}\text{Ni}(^{50}\text{Cr}, \alpha 4n)^{100}\text{Sn}$ ($\sigma = 40$ nb, experimental value) [102]. Taking into account the cross section, an estimated primary-beam intensity of $6 \cdot 10^{12}$ particles per second and an estimated number of target nuclei (assumed target thickness is 500 $\mu\text{g}/\text{cm}^2$), the production rate for ^{100}Sn can be estimated. The production rates for the mentioned reactions are around 1.1 ^{100}Sn per second and 1.2 ^{100}Sn per second, respectively.

Table 7.1: The cross sections for production of ^{100}Sn in IF fragmentation (GSI [96] and RIKEN [104]) and fusion-evaporation reaction (GANIL [102]).

Experiment	Reaction	Cross section [barn]
GSI FRS	1 GeV/u ^{124}Xe beam on Be target	$5.8 \cdot 10^{-12}$
RIKEN BigRIPS	345 MeV/u ^{124}Xe beam on Be target	$8.7 \cdot 10^{-13}$
GANIL	5.1 MeV/u ^{50}Cr beam on ^{58}Ni target	$40 \cdot 10^{-9}$

By assuming the minimum transmission through SHIP (see Section 3.1) which is 20%, rates of around 800 and 860 ^{100}Sn per hour can be expected at the focal plane of SHIP, respectively. Since SHIPTRAP has already been used to study nuclei with much lower production yields, these rates make it possible to perform a very high-precision mass measurement.

The cross sections of selected exit channels for the reaction $^{58}\text{Ni}+^{50}\text{Cr}$ in which ^{100}Sn and its neighboring nuclei are produced and the expected rates in the focal plane of the velocity filter SHIP are presented in Tab. 7.2. The production cross sections of the isobaric nuclei Ag, Cd and In are much higher than that of ^{100}Sn . In this case, mass separation is especially challenging since the relative mass difference of these isotopes with respect to ^{100}Sn is on the order of $10^{-4} - 10^{-5}$. Therefore, the background becomes the main challenge for a high-precision mass measurement of ^{100}Sn using the Penning trap technique. This can be solved by introducing an additional step of separation downstream of SHIP which is described in the next section.

Table 7.2: The cross sections for production of Ag, Cd, In and Sn isobars through $^{58}\text{Ni}+^{50}\text{Cr}$ reaction [102] together with expected rates in the focal plane of SHIP. The relative difference in mass with respect to ^{100}Sn indicates challenges in the separation of isobars.

Isobar	Cross section [barn]	Expected rate [events/s]	$\delta m = \frac{m-m_{^{100}\text{Sn}}}{m_{^{100}\text{Sn}}}$
^{100}Ag	$3.9 \cdot 10^{-3}$	27104	$2.2 \cdot 10^{-4}$
^{100}Cd	$8 \cdot 10^{-4}$	5560	$1.8 \cdot 10^{-4}$
^{100}In	$4 \cdot 10^{-6}$	28	$7.6 \cdot 10^{-5}$
^{100}Sn	$40 \cdot 10^{-9}$	0.22	

7.3 Separation of ^{100}Sn

After the reaction of primary-beam particles, which are delivered from the UNILAC with the target nuclei, the reaction products recoil out of the target. Within the velocity filter SHIP (see Section 3.1), slower reaction products are kinematically separated from the faster primary-beam particles and projectile-like reaction-products. SHIP was not designed to perform separation of ions with similar masses and isobaric nuclides. According to Tab. 7.2, this means that there will be around 10^5 times more background species passing SHIP than ^{100}Sn . Such a high amount of isobaric contaminants inside the preparation trap would introduce frequency shifts deteriorating the resolving power of the buffer-gas cooling technique (see Section 2.4.2). Therefore, in order to measure the mass of ^{100}Sn at the SHIPTRAP facility, it is required to introduce an additional separation step.

Chemical separation via molecular ions was used to produce and separate tin isotopes in the isotope separation on-line (ISOL) technique. CS_2 vapour was introduced to form SnS which is then separated from silver, indium and cadmium and ionized by an ion source for nuclear physics experiments [106]. Since this approach is not feasible at the velocity filter SHIP, a novel chemical isobaric-separation technique downstream of SHIP is proposed. Chemical reactions can take place in a gas catcher

that is usually filled with helium or argon to slow down the nuclear reaction products (see Section 2.2). A similar approach has been already applied at the LEBIT Penning trap spectrometer for mass measurements of ^{32}Si , ^{33}Si , ^{34}P and ^{32}S [107]. As the isobaric contaminant O_2^+ , that was present as an impurity, had a much higher rate than $^{32}\text{Si}^+$, water vapor was introduced into the gas catcher to form molecular ions such as $^{32}\text{SiOH}(\text{H}_2\text{O})_2^+$. The water adducts were removed by collision-induced dissociation in the RFQ cooler. The mass of $^{32}\text{SiOH}^+$ could thus be measured with a high precision to determine the mass of $^{32}\text{Si}^+$.

Normally, one of the important requirements for the operation of a gas catcher is to reduce the level of impurities as much as possible to prevent ion losses due to various reaction mechanisms (see Section 2.2.3). The quest for purity is the reason why the current SHIPTRAP gas catcher is operated at cryogenic temperatures. Unlike the cryogenic gas catcher, the main idea in the chemical-separation approach is to use a new gas catcher at room temperature and introduce chemically-reactive gases in addition to helium buffer gas. Then ^{100}Sn can be separated by utilizing its chemical properties that differ from ^{100}Ag , ^{100}Cd and ^{100}In .

Reactions of atomic cations with CH_4 in a helium environment have already been studied in Ref. [108]. The authors showed that Ag^+ and Cd^+ react with CH_4 in the following way

with the reaction-rate coefficients on the order of $k = 1 \cdot 10^{-13} \text{ cm}^3\text{s}^{-1}$. According to Ref. [108], molecular ions of In^+ and Sn^+ with methane have not been observed. Nevertheless, the ideal chemical system for the separation of Sn from its isobars is still unknown. The reaction kinetics for Sn, Ag, Cd and In are under investigation in order to find the most efficient chemical system for the separation of Sn.

The concept of ion-chemical separation of isobars at SHIPTRAP is schematically presented in Fig. 7.3. Methane is taken as an example of the chemical reagent. Ions of tin and indium, as well as molecular ions such as $(\text{CH}_4)\text{Ag}^+$ and $(\text{CH}_4)\text{Cd}^+$ are extracted from the gas catcher to an ion guide. From there, they are transferred to the following section where mass separation of Sn^+ and In^+ from $(\text{CH}_4)\text{Ag}^+$ and $(\text{CH}_4)\text{Cd}^+$ can be done by using a RFQ mass analyzer. By scanning the operating parameters of the RFQ mass analyzer, it is possible to filter out molecular ions like $(\text{CH}_4)\text{Ag}^+$ and $(\text{CH}_4)\text{Cd}^+$ (for details see Section 2.3) and at the same time transfer ions of Sn^+ and In^+ to the RFQ buncher where a continuous ion beam is converted into ion bunches in order to enable an efficient injection into the purification trap .

The purification trap allows isobaric purification of ions by applying the side-band cooling technique that is described in Section 2.4.2. To separate ^{100}Sn from ^{100}In , a resolving power higher than $R = \nu_c / \Delta\nu_c = 2.6 \cdot 10^4$ is required. A resolving power of $6 \cdot 10^4$ has already been achieved at SHIPTRAP using a cyclotron excitation time of 200 ms during mass measurements in the tin region [92]. Therefore, at this point it is possible to separate ^{100}Sn from ^{100}In and transfer it to the measurement trap for a frequency determination.

To develop the presented ion-chemical separation technique, a new setup has been built at the University of Groningen. The setup will be used for experiments to identify the most suitable chemical system for the separation of ^{100}Sn from its isobars. Once the optimum conditions for the separation are found, parts of the setup will be implemented into the SHIPTRAP facility for an accelerator-based experiment.

Figure 7.3: Schematic view of the SHIPTRAP setup that is modified for ion-chemical separation of isobars. The major components are the gas catcher and the ion guide for extraction and transfer, the RFQ mass analyzer for separation, the buncher for ion preparation and further transfer to the Penning traps. The red arrows indicate trajectories of the transferred species while the black arrows show trajectories of the separated ones.

7.4 Chemical Isobaric Separation setup

The CISE (Chemical Isobaric SEparation) setup is built for the development of ion-chemical methods for the separation of isobaric nuclei, in this particular case, the separation of ^{100}Sn and ^{100}In from ^{100}Ag and ^{100}Cd . The CISE setup consists of a gas catcher and an ion guide which is coupled to a quadrupole Time-of-Flight (qToF) mass spectrometer from Micromass UK Limited. The layout of the CISE setup is shown in Fig. 7.4.

The gas-catcher chamber is filled with helium and a reactive gas at a total pressure of around 50 mbar at room temperature. The main components of the gas catcher are the ring electrodes and a RF carpet. Stable ions of different elements inside the gas catcher can be produced for test purposes by irradiating a solid target with focused, high-power laser pulses that are delivered by a Nd:YAG laser. Depending on the type of the ablated ions and the introduced reactive gas, chemical reactions can take place inside the gas catcher resulting in the formation of neutral species or molecular ions. The behavior of the ions inside the gas catcher is described in Section 2.2 and more details on the technical design of the CISE gas catcher are given in Chapter 8.

The charged species are extracted from the gas catcher to the ion guide. The ion-guide section comprises of two hexapoles consisting of six parallel stainless-steel rods. The principle of operation is similar to that of RFQ ion guide already introduced in Section 2.3. Compared to the RFQ, the resulting potential distribution is relatively flat around the centerline which results in better ion acceptance and transmission. Hexapole 1 is placed behind the gas catcher and it is separated through a diaphragm from the second hexapole. The pressure in the first hexapole is around $4 \cdot 10^{-3}$ mbar and to prevent losses of the ions due to collisions with gas it is shorter than the second hexapole. The differential pumping reduces the pressure in the hexapole 2 to $6 \cdot 10^{-5}$ mbar. The simulation studies regarding the transport of mono-atomic ions (e.g. Ag^+) and molecular ions such as AgCH_4^+ from the gas catcher through the ion guide were performed and the details can be found in Ref. [109].

Figure 7.4: The layout of the CISE setup consisting of a gas catcher and an ion guide that are coupled to a commercial qToF. The ion guide system consists of hexapole 1 and hexapole 2. The qToF is a quadrupole mass spectrometer (Quadrupole MS) coupled with a reflection Time-of-Flight mass spectrometer (ToF MS). Figure adopted from Ref. [109].

The second hexapole is directly coupled to the MS1 that is a quadrupole mass spectrometer (Quadrupole MS). By applying a particular set of parameters one can select to operate the quadrupole analyzer as an ion guide or a mass filter (see Section 2.3). The quadrupole analyzer can be used as a mass spectrometer for ions with masses between 10 and 4000 Dalton (mass-to-charge ratio) with a mass resolving power $m/\delta m = 100$. The hexapole collision cell is used for molecular break-up studies or gas-phase studies and is followed by a hexapole transfer lens that is used for the extraction of ions and molecules. The pressure in the MS1 section is on the order of 10^{-5} mbar.

The MS2 section contains a Time-of-Flight Mass Spectrometer (ToF MS) that is operated at a pressure of 10^{-7} mbar. Depending on the voltage applied to the pusher, the ions are either directly detected at the end of the MS1 with the dynolite-point detector or extracted orthogonally into the ToF tube, where their time of flight is registered using a MCP detector. The mass-to-charge ratio m/q of the ions is identified by their ToF.

The design of the CISE gas catcher

The CISE (Chemical Isobaric SEparation) setup, introduced in Section 7.4, is used to develop an ion-chemical separation technique. After the optimum chemical system for the separation of tin from cadmium, silver and indium is established, parts of the setup will be transferred to the SHIPTRAP facility (see Chapter 3) for mass-measurements. The gas-catcher device is a part of the CISE setup and it will be utilized for offline developments as well as for accelerator-based experiments at SHIPTRAP.

In accelerator-based experiments, gas catchers play a crucial role in the preparation of ions for low-energy, high-precision measurements. The physical processes that affect the ions during slowing down and extraction are introduced in Section 2.2. In general, chemical reactions inside a gas catcher are avoided in order to prevent charge-exchange reactions that can lead to ion losses. This is the reason why special care is taken to improve the gas-purity and partially led to the development of second-generation gas catchers that operate at cryogenic temperatures [23,29].

The unique feature of the CISE gas catcher is based on the gas-phase chemistry whose possible benefits have not been investigated. The technical design of the CISE gas catcher together with the ion guide consisting of two hexapoles that are used for ion extraction is shown in Fig. 8.1. The setup is coupled to a qToF mass spectrometer (for details see Section 7.4). The gas catcher consists of the vacuum chamber and the vacuum system, the gas-supply system and the electrode system. The electrode system consists of DC ring electrodes and two RF carpets, where one RF carpet is optional, as will be discussed below. To produce ions, the ablation targets are implemented within the gas volume. The mentioned components are presented in Fig. 8.1 and will be described in the following sections.

Figure 8.1: The technical design of the gas catcher and the ion guide coupled to it. Figure adopted from Ref. [109].

8.1 Vacuum chamber

The size of the vacuum chamber is mainly based on the stopping distribution of the fusion-evaporation residues in the mass region around $A = 100$ and technical boundary conditions for the coupling of the gas catcher to the SHIPTRAP setup.

The ions from SHIP pass an entrance window made of a few micrometer thin titanium foil. The TRIM code [18] was used to simulate the energy loss of the ions through the entrance window of the gas catcher and the buffer gas inside the chamber with the aim of estimating the overall stopping efficiency. The dimensions of the inner chamber of the cryogenic-gas stopping cell of SHIPTRAP (see Section 3.2) were used in this simulations, while different settings for the ion energy up to 100 MeV, the entrance window thickness and the He pressure inside the gas catcher were considered. The simulated stopping efficiency, which is defined as the ratio of the number of ions stopped inside the gas catcher volume and the total number of ions entering the Ti window were optimized to reach 95% by selecting the appropriate thicknesses of the entrance window and the gas pressure.

The vacuum chamber of the CISE gas-catcher is shown in Fig. 8.1. It has a length of 450 mm and a diameter of 400 mm. It is designed in a way that, if needed, it can be easily divided into two subsequent sections, a 300 mm-long stopping chamber and a 150 mm-long reaction chamber. As discussed in Section 2.2.3, a large rate of ions entering the gas catcher can cause formation of plasma or molecular break-up and to avoid these problems, the first chamber can be used to slow down the ions in helium and in the following section reactive gases can be introduced.

The vacuum chamber is made from stainless steel and it contains only CF (ConFlat) flanges to ensure UHV (Ultra-High Vacuum) conditions. The stopping chamber contains a CF400 flange with an additional CF160 flange where an entrance window will be mounted in the future. The reaction chamber has a CF400 flange with electrical feedthroughs

and a CF160 connection to the following section. At the bottom of the reaction chamber, there is a CF100 flange with a connection to a turbomolecular pump. The smaller flanges (CF40 and CF25) are introduced to accommodate four vacuum-diagnostics elements and the bypass line between the two chambers, two mass-flow controllers, two mechanical feedthroughs, and two laser view ports.

8.2 The vacuum and the gas-supply system

A roots pump Leybold, ECODRY 65 plus with a pumping speed of $55 \text{ m}^3/\text{h}$ together with an Edwards, nXDS6i dry scroll pump ($6 \text{ m}^3/\text{h}$) are used to ensure rough-vacuum conditions inside the gas catcher and the ion guide consisting of two hexapoles (see Fig. 8.1). To provide high-vacuum conditions inside the gas catcher, a turbomolecular pump Leybold, TURBOVAC 450i with a pumping speed of 400 l/s for helium gas is installed at the reaction chamber. The bypass line between the stopping and the reaction chamber is introduced to evacuate the stopping chamber in the case that it is separated by an optional RF carpet, as shown in Fig. 8.1. During the operation of the gas catcher, the turbomolecular pump is separated from the vacuum chamber by a DN100 UHV gate valve. The gas catcher operates at $T = 293 \text{ K}$ and a helium pressure on the order of $p_0 = 50 \text{ mbar}$. The resulting mass-flow rate Q through the exit hole of the RF carpet with a diameter $d = 0.45 \text{ mm}$ towards the ion guide can be calculated according to the following formula [110]

$$Q = 14.2 \frac{d^2}{\sqrt{mT}} p_0 [\text{mbar l/s}], \quad (8.1)$$

where m denotes the atomic mass of the gas. From Eq. 8.1, a flow rate of helium gas ($m = 4$) into the ion guide chamber is calculated to be on the order of 4 mbar l/s . To reduce the pressure in the region of the first hexapole to a level of 10^{-3} mbar , which is needed in order to avoid ion losses and enable high-vacuum conditions in the following chambers,

a Leybold, MAG W 2200 iP turbomolecular pump with a pumping speed of 2300 l/s is used, resulting in a pressure of $2 \cdot 10^{-3}$ mbar. The vacuum conditions are monitored and controlled via full-range pressure sensors and capacitive sensors. The full-range pressure sensors Leybold, PTR 90N are used for the low-pressure measurement up to 1 mbar. The capacitive sensors Leybold, DI200 are used to control and monitor pressure inside the stopping and the reaction chamber during the operation of the gas catcher.

To perform measurements with the CISE gas catcher, it is necessary to introduce a chemical reagent in addition to the ultra-pure helium gas (typically 5.6 or 6.0). Thus, two gas lines made from stainless steel with a diameter of 6 mm are used to transfer gas to the gas catcher volume. Figure 8.1 shows the gas inlets which are located such that in the case the chamber is divided into two, the helium gas inlet will be part of the stopping chamber, while the reactive gas will be located in the reaction chamber. The gas flow of helium and the reactive gas into the gas catcher are regulated manually by a Leybold valve, UD040HR. An external mixing chamber is implemented such that it is possible to introduce several reactive gasses and adjust their concentrations by using the mass-flow controller MKS 1179A which is connected to a MKS 247C 4-channel readout. A LabView program is developed to read and control the gas flow and the pressure inside the mixing chamber [111].

8.3 Electrode system and electronics

The electrode system of the CISE gas catcher is shown in Fig. 8.2 and it comprises of DC electrodes for ion acceleration and an RF carpet which provides DC and RF fields for focusing and extraction of ions from the gas catcher.

8.3.1 DC ring electrodes

The first DC electrode contains a grid made out of wire with a diameter of 1 mm forming a mesh of 1 cm×1 cm in order to provide parallel

Figure 8.2: The DC ring electrodes and the RF carpet mounted at the extraction side of the gas catcher.

equipotential lines that will guide the ions along the axis of the gas catcher. The first 13 electrodes, when counting from the grid electrode side, are 280 mm in diameter, followed by the 14th, 15th and 16th electrodes which are 270 mm, 260 mm and 250 mm in diameter, respectively in order to match the size of the last ring of the RF carpet that is 250 mm in diameter, and thus properly shape the electric field in that region. All DC electrodes are 5 mm thick, the edges are rounded off to a radius of 2 mm and the distance between electrodes is 25 mm. The outer diameter of each DC ring electrode is 330 mm. From the outside, the DC electrodes are attached to four supporting rods that are mounted on the CF400 flange on the extraction side of the vacuum chamber. The inner part of the supporting rods is made from stainless steel, 8 mm in diameter, that is coated with 2 mm thick PEEK insulator.

A potential gradient, that has to be applied to the DC electrodes in order to guide thermalized ions is simulated with the COMSOL Multiphysics modeling software [112]. The results are shown in Fig. 8.3. The gaps between the supporting rods and the ring electrodes correspond to the PEEK insulators that are omitted in the simulation, while the stainless-steel part of the rods is set to ground. The ion motion through

Figure 8.3: DC potential distribution applied to the DC ring electrodes (left) and the resulting ion trajectories (right).

the gas catcher filled with 50 mbar of He and a DC gradient of 7 V/cm is simulated using a viscous drag model (see Section 2.2). The resulting ion trajectories are shown in Fig. 8.3, right.

8.3.2 RF carpet

As shown in Fig. 8.2, the RF carpet, that is a printed circuit board with concentric ring electrodes, is mounted at the extraction side of the gas catcher, at a distance of 15 mm from the last DC ring electrode. The outer diameter of the RF carpet is 280 mm. The design of the carpet was taken from the cryogenic stopping cell that was designed and constructed at the University of Groningen, KVI-CART and is used in FRS experiments at GSI, Germany [29, 113]. It consists of 500 concentric ring electrodes arranged over a diameter of 25 cm, with an electrode gap of 0.125 mm. The exit hole is located in the center of the RF carpet and has a diameter of 0.45 mm. The photos of the front and the back side of the carpet, together with a zoomed view of the exit hole are shown in Fig. 8.4.

Figure 8.4: The electrodes are printed on the front side of the RF carpet with a gap of 0.125 mm and a maximum diameter of 250 mm. The zoomed view of the exit hole (0.45 mm in diameter) is shown. The electronic components are placed on the back side of the RF carpet.

To transport and focus the ions towards the exit hole, a DC gradient and a RF voltage, alternating by π between adjacent electrodes, are applied to the electrodes. Due to the pressure difference between the gas catcher and the ion guide (see Fig. 8.1), the ions are carried by a gas flow through the exit hole and injected into the first hexapole. Thus, to simulate the ion trajectories, electric-field and gas-flow simulations have to be included. The left panel of Fig. 8.5 shows the resulting DC and RF potential distribution in the vicinity of the exit hole. The DC gradient set on the RF carpet electrodes is 3 V/cm, while the amplitude of the RF voltage is 100 V_{pp} with a frequency of 5 MHz. In the gas-flow simulation, a pressure of 50 mbar inside the gas catcher and 0.01 mbar in the hexapole section close to the exit hole are considered. The velocity distribution of the helium gas in the region of the exit hole is shown in Fig. 8.5, right. The trajectories of the ions in the vicinity of the exit hole calculated with a viscous-drag model are shown in Fig. 8.6.

Figure 8.5: Simulated DC and RF potential distribution (left) and the velocity distribution (right) in the region of the exit hole. These values are used as inputs to simulate ion trajectories.

Figure 8.6: The ion trajectories showing focusing and extraction of ions in the region of the exit hole.

8.3.3 Electronics

The conceptual design of the electronics system and the wiring is shown in Fig. 8.7. To create the DC gradient at the DC ring electrodes, a maximum voltage, DC_{max} , is applied to the first DC electrode containing the grid, while a minimum voltage, DC_{min} , is applied to the DC electrode closest to the RF carpet. The DC electrodes are connected by a chain of $50\text{ k}\Omega$ resistors to form a potential gradient. The voltages are generated by a high-voltage power supply from iseg with a maximum voltage output up to 2 kV .

Figure 8.7: DC and RF voltage arrangement. The ring electrodes are connected by a resistor chain and supplied with a DC_{max} and a DC_{min} voltage to create the DC gradient. The same is valid for the RF carpet electrodes which are additionally supplied with the RF voltage with opposite phases.

The same power supply is used to provide a DC gradient at the RF carpet electrodes. The DC voltage is arranged similarly, with a $DC_{\max}$ applied to the outer electrode and a $DC_{\min}$ applied to the electrode that is closest to the exit hole. The gradient is created by a voltage divider that is integrated on the back side of the RF carpet (see Fig. 8.4).

To supply the RF carpet with the alternating voltage, the sinusoidal function of the RF voltage is delivered by a signal generator Keysight 8656B. In order to provide higher amplitudes, typically around $100 V_{pp}$ at frequencies up to 10 MHz, the signal is amplified by an RF power amplifier RM Italy KL505 and drives a resonance circuit that consists of a 160 nH-inductor coupled in parallel to the capacitance of the ring that is 6 nF and two 940 pF capacitors in series to match the circuit to the 50Ω source at a specific resonance frequency. The RF voltage reaches a maximum value of $100 V_{pp}$ at around 5 MHz. The RF signal is capacitively coupled to the rings such that neighboring rings have opposite RF phases, RF_+ and RF_- . The design of the RF carpet electronic circuit is adopted from Ref. [29]. The electronic circuit that combines an RF phase with the DC voltage is integrated on the backside of the carpet and can be seen in Fig. 8.4.

8.4 Laser-ablation ion source

The laser-ablation ion source is introduced in order to produce stable ions and investigate their gas-phase chemistry within the gas catcher. A frequency-doubled Nd:YAG laser at 532 nm (Continuum, Minilite series) is employed to irradiate a target sample and produce positive ions from the surface material. The energy of the laser pulse is adjustable up to 25 mJ, while the pulse length is 5 ns at a maximum repetition rate of 15 Hz. In order to increase the power density on the target, the laser beam is focused using optical lenses.

Two target holders are mounted on the mechanical feedthroughs Linear Power Probe from UHV design and can be moved through the DC electrodes. Each target holder contains space to mount four targets with

Figure 8.8: A target holder with four different targets mounted. Each target can contain multiple samples for the production of several ion species simultaneously.

thicknesses varying between 0.25 mm to 3 mm and a size of 17.5×12 mm, and if needed, can contain multiple samples as shown in Fig. 8.8. This is convenient for gas-phase chemistry investigations as different species can be produced at the same time. The samples can be selected either by moving vertically the target holder with the samples or by manually moving the laser beam by optical elements in order to select different samples on the same target. Production of ions at the two different positions with respect to the longitudinal axis of the gas catcher enables investigating chemical reaction yields depending on the different extraction times. If the main chamber is divided by an RF carpet into two subsequent chambers, both can be tested and optimized by using the laser-ablation ion source.

8.5 Proof of ion extraction

The described components of the gas catcher are built and assembled at the University of Groningen. The gas catcher is coupled to the qToF

mass spectrometer via the double hexapole ion guide. The setup is shown in Fig. 8.9 and the details concerning the ion guide and the qToF are given in Section 7.4.

Figure 8.9: The gas catcher device coupled to the qToF.

Figure 8.10: ToF spectrum of the cadmium isotopes extracted from the gas catcher. The ions are produced by irradiating the cadmium target using Nd:YAG laser at 532 nm and 80 mW laser power.

To perform off-line tests, the gas catcher was filled with 50 mbar of helium. A DC gradient was applied to the electrodes of the DC cage ($DC_{\max} = 297\text{ V}$ and $DC_{\min} = 18\text{ V}$) and the RF carpet ($DC_{\max} = 39\text{ V}$ and $DC_{\min} = 0\text{ V}$). The RF carpet was operated at its resonance frequency of 5.2 MHz and the voltage of 166 V_{pp} . The cadmium sample was mounted on the movable target holder which was biased at 40 V. The laser-ablated cadmium ions are extracted from the gas catcher and transferred to the qToF where their ToF distribution was recorded by using a MCP detector. The obtained spectrum containing six stable isotopes of cadmium is shown in Fig. 8.10. The peak heights reflect the natural abundance of cadmium.

To investigate the extraction time and the extraction efficiency, a set of measurements using laser-ablated ions and a radioactive ion source will be performed. Once the optimum operating parameters are fixed, the reactive gas will be introduced and the investigation of the reaction kinematics will be performed with laser-ablated ions of tin, indium, cadmium and silver.

Summary and outlook

This thesis presents high-precision mass measurements of the heaviest nuclides performed with the SHIPTRAP mass spectrometer, as well as technical developments directed towards studies of medium-heavy neutron-deficient isotopes in the region around ^{100}Sn .

Direct mass measurements of very heavy and superheavy nuclides provide important quantities, such as nucleon separation energies and shell gaps, which are used to map the strength of shell effects. These are crucial for nuclear models attempting to pinpoint the position of the predicted island of stability. So far, direct mass measurements were performed up to the region of nobelium and lawrencium isotopes. Accurate mass measurements in this region can serve as anchor points to indirectly determine the atomic masses of heavier nuclei.

Motivated by the success of pioneering experiments performed in 2010 at SHIPTRAP in the region of the heaviest elements [5–7], a long effort to boost the efficiency of the setup has been undertaken in order to step towards even more exotic species. During the experimental campaign in 2018, high-precision mass spectrometry of $^{251,254}\text{No}$ and $^{254-256}\text{Lr}$ was performed at SHIPTRAP by employing the PI-ICR technique. For the

first time, the atomic ground-state masses of the odd- A isotope ^{251}No and the odd-odd isotope ^{254}Lr have been measured directly as well as the ground-state mass of the superheavy nucleus ^{257}Rf . In addition, low lying isomeric states were resolved for the first time with Penning trap mass spectrometry at the extreme of low count rate and low number of events. The excitation energies of the long-lived, low-lying isomeric states $^{251\text{m},254\text{m}}\text{No}$ and $^{254\text{m},255\text{m}}\text{Lr}$ have been determined. Within this work, the ground-state masses were analyzed and relative mass uncertainties down to $\delta m/m = 10^{-9}$ were obtained. The results are in agreement with the values from the atomic mass evaluation 2016 [81] and confirm the strength of the deformed shell closure at $N = 152$. An overview of the measured masses in the region above uranium is shown in Fig. 9.1 together with the results obtained in this work. In combination with decay spectroscopy studies, our results will indirectly improve the atomic masses of isotopes up to the element darmstadtium ($Z = 110$) which will contribute to the investigation of the strength of the shell closure at $N = 162$.

This remarkable achievement has been accomplished thanks to the implementation of the cryogenic gas-stopping cell, which boosted the overall efficiency of the SHIPTRAP setup to about 5%, and due to the development of the PI-ICR technique, which is characterized by high precision, supreme mass resolving power and fast measurements. During the beamtime in 2020, the efficiency of the cryogenic gas-stopping cell and the RFQ buncher was further optimized for ^{257}Rf . Mass measurements could not be performed at that time due to technical problems with the Penning traps. However, the ground and the isomeric states of $^{255,257}\text{Rf}$ will be addressed in the experimental campaign in 2021, together with one of the Db ($Z = 105$) isotopes. In parallel to these activities, efforts focused on adapting the non-destructive Fourier-Transform Ion-Cyclotron-Resonance technique [41] to the region of superheavy elements are ongoing. A second dedicated Penning-trap setup (SHIPTRAP 2) is under development and will be coupled to the existing beam line of SHIPTRAP. This technique will allow direct mass measurements of a single ion with half-lives above 1 s and enable studies of

Figure 9.1: Part of the nuclear chart in the region of the heaviest elements. Blue squares correspond to the nuclei the masses of which have been measured directly within this work. These results in combination with decay spectroscopy studies will allow indirect mass determination of the heavier isotopes displayed by green. The isotopes displayed by magenta, orange and red have been measured directly at TRIGATRAP [114], RIKEN [84] and SHIPTRAP [5, 6], respectively. The dashed lines show experimentally established deformed shell closures while the solid lines indicate the location of the island of stability predicted by macroscopic microscopic models. Figure modified from Ref. [115].

heavier elements with lower production cross sections.

Aside from investigations in the region of the heaviest elements, SHIPTRAP has been used in the mass measurements of medium-heavy neutron-deficient isotopes near the $N = Z$ line [92]. Here, high-precision mass spectrometry of doubly-magic ^{100}Sn is of particular interest not only for nuclear structure studies, but also for a better understanding of the astrophysical rp-process [8]. In this work, the main challenges concerning mass spectrometry of ^{100}Sn at SHIPTRAP are discussed. A novel

chemical separation technique is proposed to separate ^{100}Sn from its isobars by taking advantage of gas-phase chemistry inside a gas catcher. To develop this technique, a new gas catcher was designed and it has been built. It will be used as a part of the CISE (Chemical Isobaric Separation) setup to systematically study the gas-phase chemistry of tin, silver, cadmium and indium. The commissioning of the setup with the gas catcher and an ion guide connected to a qToF mass spectrometer is ongoing. Once the optimum conditions for the separation of tin are established, the gas catcher will be implemented at SHIPTRAP to measure the atomic mass of ^{100}Sn and isotopes in its vicinity with the highest precision.

Nederlandse samenvatting en vooruitzicht

Dit proefschrift presenteert hoge-precisie massametingen van de zwaarste nucliden uitgevoerd door de SHIPTRAP massaspectrometer, evenals technische ontwikkelingen gericht op onderzoeken van middelzware neutron-arme isotopen in de regio rond ^{100}Sn .

Directe massametingen van zeer zware en superzware nucliden bepalen belangrijke concepten zoals nucleon scheidingsenergiën en “shell gap”, die gebruikt worden om de kracht van schileffecten in kaart te brengen. Deze zijn cruciaal voor nucleaire modellen om de positie van het voorspelde eiland van stabiliteit aan te wijzen. Tot dusver zijn directe massametingen uitgevoerd voor isotopen tot en met de regio van nobelium en lawrencium. Nauwkeurige massametingen in deze regionen kunnen dienen als referentiepunten om de massa van zwaardere kernen indirect te kunnen vaststellen.

Gemotiveerd door het succes van baanbrekende experimenten uitgevoerd in 2010 bij SHIPTRAP in de regio van de zwaarste elementen [5–7] is een grootse inspanning ondernomen om de efficiëntie van

de experimentele opstelling te verbeteren om zo de stap te maken naar metingen van nog exotischere nucliden. Tijdens de experimentele campagne in 2018 was hoge-precisie massaspectrometrie van $^{251,254}\text{No}$ en $^{254-256}\text{Lr}$ uitgevoerd bij SHIPTRAP door de toepassing van de PI-ICR (Phase-Imaging Ion-Cyclotron-Resonance) techniek. Voor het eerst waren direct de massa's gemeten van de grondtoestand van de isotoop met oneven A -waarde ^{251}No en de oneven-oneven isotoop ^{254}Lr evenals de massa van de grondtoestand van de superzware kern ^{257}Rf . Daarnaast is het gelukt om, hoewel met een extreem laag aantal meetpunten, laagliggende isomere toestanden te onderscheiden van de grondtoestand door middel van Penningval massaspectrometrie. De excitatie-energie van de langlevende, laagliggende isomere toestanden $^{251\text{m},254\text{m}}\text{No}$ en $^{254\text{m},255\text{m}}\text{Lr}$ zijn bepaald. Binnen dit werk zijn de massa's van de grondtoestanden geanalyseerd en zijn relatieve massa onzekerheden tot $\delta m/m = 10^{-9}$ verkregen. Deze resultaten zijn in overeenstemming met de waarden van de atomaire massa evaluatie uit 2016 [81] en bevestigen de sterkte van de vervormde schilsluiting op $N = 152$. Een overzicht van de gemeten massa's in de regionen boven uranium is te zien in Fig. 9.1 samen met de resultaten die verkregen zijn met dit werk. In combinatie met vervalspectroscopieonderzoeken zullen onze resultaten indirect de atomaire massa van isotopen tot en met element darmstadtium ($Z = 110$) verbeteren, wat zal bijdragen aan onderzoek van de sterkte van de schilsluiting bij $N = 162$.

Deze opmerkelijke prestatie is volbracht dankzij de implementatie van een cryogene gasstopcel, welke de efficiëntie van de SHIPTRAP opstelling ongeveer 5% verbeterde, en door de ontwikkeling van de PI-ICR techniek, welke is gekarakteriseerd door hoge precisie, fantastisch massascheidend vermogen en snelle metingen. Tijdens de meetperiode in 2020 zijn de efficiëntie van de cryogene gasstopcel en de RFQ bundelaar verder geoptimaliseerd voor ^{257}Rf . Massametingen konden op dat moment niet uitgevoerd worden vanwege technische problemen met de Penningvallen. Echter, de isomere en grondtoestanden van $^{255,257}\text{Rf}$ zullen behandeld worden tijdens de experimentele campagne in 2021, samen met een van de Db ($Z = 105$) isotopen. Parallel aan deze activiteiten wordt

Figuur 10.1: Deel van de nuclidenkaart in de regio van de zwaarste elementen. De blauwe vierkanten komen overeen met de nucliden waarvan de massa is gemeten binnen dit werk. Deze resultaten zullen samen met vervalspectroscopieonderzoeken de indirecte massabepaling toestaan van zwaardere isotopen weergegeven in het groen. De isotopen in het magenta, oranje en rood zijn direct gemeten door respectievelijk TRIGATRAP [114], RIKEN [84], en SHIPTRAP [5, 6]. De stippellijnen tonen experimenteel vastgestelde vervormde schilsluitingen terwijl de ononderbroken lijnen de locatie aanduiden van het eiland van stabiliteit voorspeld door macroscopische-microscopische modellen. Figuur gewijzigd van Ref. [115].

de aandacht gefocust op het aanpassen van de niet-destructieve FT-ICR (Fourier-Transform Ion-Cyclotron-Resonance) techniek [41] aan de regionen van superzware elementen. Een tweede Penningvalopstelling toegewijd hieraan (SHIPTRAP 2) is in ontwikkeling en zal worden gekoppeld aan de bestaande straallijn van SHIPTRAP. Met deze techniek kunnen directe massametingen gedaan worden van enkele ionen met een halfwaardetijd van boven 1 seconde en zal het mogelijk worden gemaakt om zwaardere elementen met lagere werkzame doorsnede te onderzoeken.

Naast het onderzoek in de regionen van de zwaarste elementen is SHIP-TRAP ook gebruikt in massametingen van middelzware, neutronarme isotopen dichtbij de $N = Z$ lijn [92]. In dit gebied is hoge-precisie massaspectrometrie van dubbel-magisch ^{100}Sn van grote interesse. Niet alleen voor het bestuderen van de nucleaire structuur, maar ook voor een beter inzicht in het astrofysische rp-proces [8]. In dit proefschrift, de belangrijkste uitdagingen met betrekking tot de massaspectrometrie van ^{100}Sn door SHIPTRAP worden besproken. Een chemische scheidingstechniek wordt voorgesteld om ^{100}Sn te scheiden van zijn isobaren, gebruik makend van gas-fase scheikunde binnenin een gasvanger. Een nieuwe gasvanger was ontworpen en gebouwd om deze techniek te ontwikkelen. Het gaat gebruikt worden als deel van de CISE (Chemical Isobaric Separation) opstelling om systematisch de gas-fase scheikunde van tin, zilver, cadmium en indium te bestuderen. Het inbedrijfstellen van de opstelling met de gasvanger en een ionenvervoerder verbonden aan een qToF massaspectrometer is lopende. Wanneer de optimale omstandigheden voor de scheiding van tin met zijn isobaren zijn bereikt, wordt de gasvanger geïmplementeerd in SHIPTRAP om de atomaire massa van ^{100}Sn en isotopen binnen die regio te meten met de hoogst mogelijke precisie.

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