

The evolution of nuclear moments and charge radii approaching ^{100}Sn

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Abstract

In this work, the electromagnetic properties of the tin atom and its nucleus are systematically investigated in order to test our description of complex many-body quantum systems, such as bound electrons in the atom, and the protons and neutrons within the nucleus.

The main motivation of this thesis is to investigate the evolution of nuclear electromagnetic properties (magnetic dipole moments, spin, electric quadrupole moments and charge radii) when neutrons are removed from the tin nucleus, towards ^{100}Sn ($N = Z = 50$). This neutron-deficient nucleus (12 neutrons away from the lightest stable tin isotope) is expected to exhibit the extraordinarily rare property of having an equal number of protons and neutrons in doubly-magic (closed proton and neutron shell) configuration. Doubly-magic nuclei are more tightly bound than their neighbouring nuclei and provide valuable stepping stones for investigating the influence on nuclear structure properties when adding or removing nucleons (protons or neutrons). The tin isotopic chain is expected to have two doubly-magic nuclei, the neutron-deficient ^{100}Sn ($N = Z = 50$) and neutron-rich ^{132}Sn ($N = 82, Z = 50$). While the doubly-magic nature of ^{132}Sn has been firmly established, the shell-closures at ^{100}Sn have not yet been confirmed due to the low production yields of the neutron-deficient tin isotopes. Interestingly, measurements hint towards an increased softness to deformation when approaching ^{100}Sn which questions the robustness of the shell closure. Additionally, the controversial level ordering of the two neutron shell model orbits above $N = 50$, labeled $2d_{5/2}$ and $1g_{7/2}$, further fuel the curiosity.

Thanks to the highly sensitive Collinear Resonance Ionization Spectroscopy (CRIS) technique at ISOLDE-CERN, the above questions could be addressed in this work. Using the CRIS technique, laser spectroscopic measurements of the hyperfine structures and isotope shifts could be resolved in atoms containing tin isotopes artificially produced at ISOLDE, ranging from ^{124}Sn down to ^{104}Sn . In preparation for these measurements, resonance ionization schemes in atoms containing the stable tin isotopes were extensively investigated.

These measurements combined with with state-of-the-art quantum chemistry calculations revealed the sensitivity of different atomic transitions in tin to the nuclear structure properties, such as the nuclear magnetic dipole and electric quadrupole moments as well as the changes in mean-square charge radii.

This work reveal that the ground-state nuclear moments of ^{105}Sn is in a good agreement with a sequential shell model filling of neutrons in the $2d_{5/2}$ orbital above ^{100}Sn . An enhanced collectivity is observed when adding neutrons in the $1g_{7/2}$ orbital, which is suggested to originate from the strong proton-neutron correlations driving proton excitations across the $Z = 50$ shell gap. Furthermore, the changes in the mean-square charge radii along the tin isotopic chain reveal a new perspective of the $N = 64$ subshell closure when filling the $2d_{5/2}$ and $1g_{7/2}$ neutron orbitals. These results are compared with state-of-the-art many-body calculations using interactions founded on the fundamental description of the strong nuclear force, only constrained by nuclear properties of light nuclei $A \leq 4$. The presented comparison between experiment and theory highlight the incredible progress as well as the current limitations in our quest towards a fundamental description of nuclear structure.

Beknopte samenvatting

In dit werk worden de elektromagnetische eigenschappen van het tin atoom en in het bijzonder zijn atoomkern systematisch onderzocht om de huidige theoretische beschrijving van complexe veeldeeltjes-quantumsystemen, zoals de gebonden elektronen in het atoom, en de protonen en neutronen in de kern, te testen.

De belangrijkste motivatie van dit proefschrift is het onderzoeken van de evolutie van nucleaire elektromagnetische eigenschappen (magnetische dipoolmomenten, spin- en elektrische quadropoolmomenten en ladingsstralen) wanneer neutronen systematisch worden verwijderd uit de tin kern, tot we de zwaarste bekende kern met een gelijk aantal protonen en neutronen bereiken: ^{100}Sn ($N = Z = 50$). Deze neutronen-arme kern (met 12 neutronen verwijderd van de lichtste stabiele tin kern) heeft naar verwachting gesloten proton- en neutronenschillen in een zogenaamde dubbel-'magische' configuratie en is daarom bijzonder waardevol als opstap in het nucleaire landschap voor het bouwen van een de theoretische beschrijving van zwaardere kernen. State-of-the-art veeldeeltjes technieken, die gebaseerd zijn op, waaronder interacties die de fundamentele sterke kernkracht beschrijven vertrekkende vanuit QCD, hebben deze belangrijke opstap in het nucleaire landschap kunnen bereiken. Er blijven echter veel vragen onbeantwoord met betrekking tot de structuur en de fundamentele eigenschappen van de lichtste tin isotopen vanwege het gebrek aan experimentele gegevens. Onder andere is de robuustheid van de $Z, N = 50$ magische getallen nog controversieel, en is er nog onvoldoende begrip over de niveau-ordening van de neutronen $2d_{5/2}$ en $1g_{7/2}$ quantum orbitalen, alsook over de collectieve eigenschappen van de nucleonen in deze kernen.

Dankzij de zeer gevoelige collineaire resonantie-ionisatiespectroscopie (CRIS)-techniek bij ISOLDE-CERN, kunnen enkele van de bovenstaande vragen in dit werk worden beantwoord door de elektromagnetische eigenschappen van tin isotopen in de grondtoestand te bestuderen tot ^{104}Sn . Ter voorbereiding van deze metingen werden resonantie-ionisatieschema's in tin onderzocht met

behulp van een off-line ablatie-ionenbron gekoppeld aan de CRIS-bundellijn, om de tien stabiele isotopen van tin te produceren en te bestuderen. Het inzicht in hun gevoeligheid aan specifieke nucleaire eigenschappen (kernstraal, spin, momenten) legde de basis voor de studie van de kortlevende tin isotopen, terwijl het waardevolle tests opleverde voor de modellen die atomaire eigenschappen voorspellen. Twee resonantie-ionisatieschema's die in dit werk zijn ontwikkeld, werden bij CRIS gebruikt voor het uitvoeren van metingen van de hyperfijne structuren van atomair tin dat de onstabiele isotopen bevat. De resultaten van deze metingen werpen nieuw licht op de niveau-ordering van de neutronenkwantumbanen en het collectieve gedrag in isotopen die ^{100}Sn naderen. Deze resultaten worden direct vergeleken met state-of-the-art nucleaire *ab-initio* berekeningen die de ongelooflijke vooruitgang en de huidige beperkingen van deze berekeningsmethoden onthullen.

List of Abbreviations

AI Auto-Ionizing. 101

Bfi Birefringent Filter. 61

CEC Charge-Exchange-Cell. 58

CRIS Collinear Resonance Ionization Spectroscopy. 4

CSV Comma-separated values. 69

CW Continuous-Wave. 61

DAQ Data acquisition. 69

eV electron volt. 31

FS Field-shift. 33

IP Ionization Potential. 47

IS Isotope shift. 35

MS Mass shift. 34

PDL Pulsed Dye Laser. 61

PSB Proton Synchrotron Booster. 53

QCD Quantum chromo dynamics. 1

QED Quantum electro dynamics. 1

RILIS Resonance Ionization Laser Ion Source. 48

S.E Schrödinger Equation. 8

Ti:Sa Titanium-Sapphire. 61

TOF Time-of-Flight. 51

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Chapter 1

Introduction

The quest for a universal description of nature, which would allow us to predict the properties of the smallest indivisible units of matter as well as the grand scale of the universe, is one of the most fundamental goals in physics. This great endeavour has resulted in the standard model, developed in the early 1970s, which describes observed phenomena in nature in terms of three (strong, weak, electromagnetic) of four known forces (not yet including gravity) together with a set of point-like elementary particles [1]. This standard model, was inspired by a mathematically elegant description of the first discovered elementary particle, the electron, from the quantization of the electromagnetic field with quantum electrodynamics (QED). Thus far, this description is the closest we have reached to a fundamental truth, which has allowed us to predict the behaviour of individual electrons with a remarkable precision [2]. The negatively charged electrons are often bound by the electromagnetic force within atoms. The understanding of bound electrons allow us to describe the chemical properties of the elements in our universe. In the heart of the atom lies the nucleus, which is composed of neutrons and positively charged protons bound against the electrostatic repulsion by the strong nuclear force. Unlike electrons, protons and neutrons are composite particles containing the elementary particles known as quarks and gluons, as described by quantum chromodynamics (QCD) [3]. While much is known at the length scale of individual quarks, a QCD based description extending to the scale of the nucleus is still missing. Only after a fundamental framework of the nuclear forces is understood, can one reach a complete description of the atomic nucleus, and its properties. This goal inspires the systematic study of properties of nuclei with different combinations of protons and neutrons. Most combinations of protons and neutrons in nuclei are short-lived and do not occur naturally on earth, making this endeavour

particularly challenging. Sophisticated experimental techniques have been developed in the last century which has allowed us to artificially create and study exotic radioactive nuclei in the far reaches of the nuclear landscape, as seen in Fig. 1.1. These measurements reveal interesting properties which continue to challenge our description of the atomic nucleus [4, 5, 6, 7, 8].

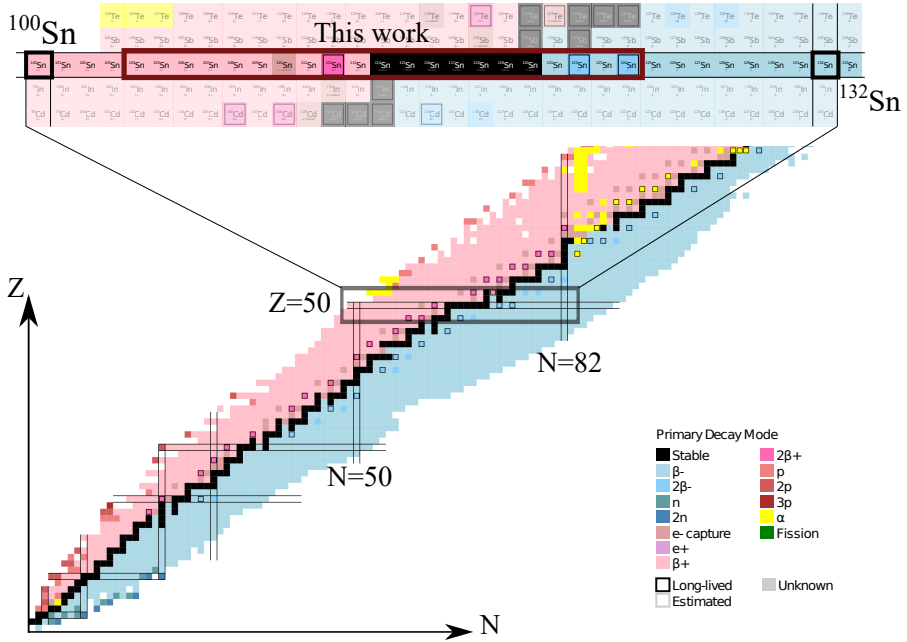


Figure 1.1: The nuclear chart highlighting the tin isotopic chain investigated in this thesis.

Despite the underlying complexity of atomic nuclei, phenomenological models are able to successfully describe many of its characteristic properties. For example, bulk or 'collective' properties such as vibrations and rotations of deformed nuclei were initially explained using the liquid drop model, formulated by George Gamow in the beginning of the 1930s [9]. Further insight was revealed from measurements of decay properties of radioactive nuclei performed in the early 20th century. These measurements revealed characteristic patterns of enhanced binding energy¹, observed at (2,8,20,28,50,82,126), with added numbers of protons or neutrons in the nucleus. These so-called 'Magic' numbers

¹Binding energy is defined as the energy required to separate all protons and neutrons from the nucleus.

were later described by the nuclear shell model from the filling of independent proton or neutron shell orbitals within the nucleus, analogous to electrons filling atomic shell model orbitals. This description of the nuclear shell model awarded the Nobel prize in physics to Maria Goeppert Mayer and J. Hans D. Jensen in 1963 [10, 11, 12]. Nuclei with both filled proton and neutron shells, known as doubly-magic nuclei are particularly rare. Only 8^2 out of over 3000 nuclei discovered so far, are confirmed to fulfill this criteria [13], as presented in Fig. 1.1.

These unique nuclei are more tightly bound when compared to their neighbouring isotopes³ and exhibit properties analogous to noble gases of the chemical elements. Consequently, doubly-magic nuclei act as unique stepping stones in the nuclear landscape, where the evolution of nuclear properties can be carefully studied when adding or removing nucleons. The element tin, having a magic number of protons ($Z = 50$), is of particular interest as it has the longest isotopic chain with two expected doubly-magic isotopes at the far ends of the nuclear landscape: The neutron-deficient ^{100}Sn ($N = Z = 50$) and the neutron-rich ^{132}Sn ($Z = 50, N = 82$), highlighted in Fig. 1.1. This isotopic chain provides an excellent testing ground for investigating the influence of added neutrons between two shell-closures situated at the extreme ends of the nuclear landscape. While the strong doubly-magic shell-closures with $Z = 50$ and $N = 82$ at ^{132}Sn have been firmly established [14, 15, 16, 17, 18], the $Z = N = 50$ shell closures at ^{100}Sn has not yet been experimentally confirmed. This nucleus is of particular importance as it is the last doubly-magic nucleus with an equal number of protons and neutrons predicted to exist in a bound state. Inconveniently, the extreme difficulty to artificially produce this exotic nucleus and its neighbouring isotopes, with half-lives of a few minutes to less than a second, means only a few measurements have been performed in its vicinity so far. Interestingly, the measured excitation probability to the first excited state of even-even tin isotopes have indicated an enhanced softness to vibrations when approaching ^{100}Sn , in contradiction to shell-model predictions [19, 20, 21]. These measurements question the robustness of the predicted doubly-magic shell closure. Furthermore, the closely spaced neutron quantum orbits above ^{100}Sn are heavily debated due to contradictory measurements suggesting different ordering [22, 23]. More measurements are thus needed in order to clarify the controversies regarding the nuclear structure evolution approaching ^{100}Sn and to test our description of the atomic nucleus at the limits of nuclear existence.

Nuclear properties such as spins, magnetic dipole moment, electric dipole moments and charge radii can be revealed by precisely studying the energies of

²With ^4_2He , $^{16}_8\text{O}$, $^{40}_{20}\text{Ca}$, $^{48}_{20}\text{Ca}$, $^{56}_{28}\text{Ni}$, $^{78}_{28}\text{Ni}$, $^{132}_{50}\text{Sn}$ and $^{208}_{82}\text{Pb}$ confirmed at the time of writing.

³Nuclei with the same number of protons (Z) but different number of neutrons (N).

the electrons bound in the atom. While magnetic dipole moments and spins are sensitive to the microscopic orbital configuration of individual protons and neutrons within the nucleus, electric quadrupole moments and charge radii provide an overarching insight into the collective nuclear properties, such as the shape and size of the nuclear charge distribution. In this respect, sensitive laser spectroscopy techniques have been established as a valuable tools for probing both the macroscopic and microscopic nuclear properties of both stable and short-lived nuclei produced at radioactive beam facilities [24].

In this thesis, a particularly sensitive laser spectroscopy technique known as the collinear resonance ionization spectroscopy (CRIS) is used as a tool for investigating the evolution of nuclear moments and charge radii approaching ^{100}Sn . In order to realize these measurements it was essential to understand the electronic configuration in the tin atom most sensitive to the nuclear properties of interest. With the combined effort of state-of-the-art quantum chemistry methods and experimental measurements of stable tin isotopes, a comprehensive overview of the sensitivity of different electronic configurations to the nuclear observables in tin was created. This work is presented in the peer-reviewed article:

1. Tin resonance-ionization schemes for atomic-and nuclear-structure studies, **Gustafsson, F. P**, Ricketts, C. M , Reitsma, M. L, Garcia-Ruiz, R. F, Bai, S.W, Berengut, J.C, Billowes, J, Binnersley, C.L, Borschevsky, A , Cocolios, TE *et al.*
Physical Review A, 102(5), 052812, 2020.

Two newly developed laser ionization-schemes developed from this work were utilized at the CRIS experiment for performing laser spectroscopy of 21 tin isotopes from ^{124}Sn down to ^{104}Sn , highlighted in Fig. 1.1. These measurements provide valuable new insights into the ground-state arrangement of neutrons occupying the two lowest lying shell model orbitals above $N = 50$ ($2d_{5/2}$ and $1g_{7/2}$). While the evolution of nuclear charge radii and electric quadrupole moments reveal a new perspective of the collective properties approaching ^{100}Sn .

Finally, the electromagnetic properties of the tin nuclei are used as benchmarks for testing state-of-the-art many-body methods founded on a theoretical framework known as chiral effective field theory (χEFT). This theoretical framework provides a link to the underlying symmetries of QCD for an *ab-initio* (first principles) description of the nucleus [25, 26]. The rapid progress of many-body calculation methods based on this *ab-initio* framework have in the recent decades been able to describe properties of increasingly more complex nuclei, which push our understanding of the fundamental nuclear forces. Recently, the tin region has been reached [27]. The results presented in this thesis highlight

the remarkable progress as well as the remaining challenges in our path to reach a unified description of the atomic nucleus.

Chapter 2

Nuclear structure and the interest in tin

In this chapter the concept of many-body quantum systems is outlined in the context of the nucleons (protons and neutrons) bound in nuclei. In order to simplify the description of this complex system the mean-field approximation is often used. The concept of independent particles bound in the mean-field potential generated by $N-1$ nucleons, form a foundation for an independent particle shell model description of the nucleus, presented in Sec. 2.2. The shell model concept is developed further by introducing interactions between individual nucleons in the interacting particle shell model, Sec. 2.3. This is followed by Sec. 2.4, which contains a brief overview of a modern *ab-initio* based description of the nucleus together with the interactions founded on χ EFT employed in this work. A general description of the nuclear electromagnetic properties of interest in this thesis is provided in Sec. 2.5. This is followed by the short summary of some of the remaining questions left unanswered regarding the nuclear structure near ^{100}Sn , which motivate this thesis work.

2.1 Many-body quantum systems

The state of a N-body quantum system is described by its wave-function $|\Psi(t)\rangle$. The total energy can be acquired when a solution is found to the eigenvalue problem of the Hamiltonian operator $\hat{H}$, commonly referred to as the time-independent Schrödinger equation (S.E.):¹

$$\hat{H}|\Psi\rangle = (\hat{T} + \hat{V})|\Psi\rangle = E|\Psi\rangle \quad (2.1)$$

where $\hat{H}$ is composed of the sum of kinetic energy $\hat{T}$ and potential energy $\hat{V}$ operators for the N-bodies system,

$$\hat{T} = \sum_i^N -\frac{\hbar^2}{2m_i} \nabla^2 \quad (2.2)$$

$$\hat{V} = \sum_{i<j}^N v_{i,j} + \sum_{i<j<k}^N v_{i,j,k} + \sum_{i<j<k<l}^N v_{i,j,k,l} + \dots \quad (2.3)$$

where m_i is the mass of the i^{th} particle in the system, $v_{i,j}$ is the potential between 2-bodies (NN) and $v_{i,j,k}$ for 3-bodies (NNN) etc. Hypothetically, if the wave-function of the system is known from the solution of Eqn. 2.1, any observable could be calculated as the expectation value of the appropriate operator. However, constructing the S.E. is particularly challenging as it requires an accurate formulation of the contributions from all N-body interactions, Eqn. 2.3. While NN interactions have been investigated through measurements of nucleon-nucleon scattering, 3-body interactions are not as well understood. Even classical 3-body systems result in chaotic solutions for most initial conditions. Non-determinism in quantum mechanics means that finding an accurate solution to N-body quantum systems is controversial if even possible at all. The interplay of the three different forces (strong, weak and electromagnetic) involved in nuclear systems is largely unknown, which further adds to the complexity of the nuclear many-body problem. Approximations are therefore required when formulating a description of the nucleus. A commonly used approach to treat this many-body problem is by assuming that each interacting body is bound in a static mean-field potential $U(i)$ generated by N-1 bodies:

¹For systems involving lighter mass particles at higher velocities the relativistic Dirac Hamiltonian is required. The non-relativistic Hamiltonian is often sufficient to treat the motion of protons and neutrons in the nucleus.

$$\left(\underbrace{\sum_{i=1}^N \left[-\frac{\hbar^2}{2m_i} \nabla_i^2 + U(i) \right]}_{\hat{H}_0} + \underbrace{\left[\hat{V} - \sum_{i=1}^N U(i) \right]}_{\hat{V}_R} \right) |\Psi(t)\rangle = E |\Psi(t)\rangle. \quad (2.4)$$

Where $\hat{H}_0$ is the independent particle Hamiltonian describing individual nucleons bound in the mean-field potential. The second term, $\hat{V}_R$, is the residual interaction which contain the leftover N-body interactions, where often only the 2-body NN interactions are considered. These residual interactions can be treated as perturbations of the mean-field Hamiltonian.

2.2 The independent particle shell model

In the beginning of the 20th century experimental evidence from precision measurements of different nuclear properties indicated a characteristic pattern of enhanced binding energy in nuclei with specific proton and neutron numbers (2,8,20,28,40,50,82,126) [10, 11, 12]. These so-called 'magic' numbers showed many similarities with the electronic shell model developed earlier to describe electrons bound by the electrostatic attraction to the positively charged nucleus. Consequently, a nuclear shell model was also formulated for describing protons and neutrons bound by the strong nuclear force within the nucleus. In this nuclear shell model description, the protons and neutrons are described as independent particles bound by each respective mean-field potential (ignoring $\hat{V}_R$ in Eqn. 2.4).

As a first guess a simple harmonic oscillator (S.H.O) potential was used $U(r) = m\omega^2 r^2/2$, where r is the orbital radius and ω is the angular frequency of the oscillator. This simple guess reproduced the magic numbers for light nuclei up to 20, but failed to describe the shell closures when adding more nucleons, see Fig. 2.1. The inclusion of an attractive orbital angular momentum term l^2 broke the degeneracy of the principal quantum levels and lowered the potential for nucleons with greater angular momentum number. Finally, by adding the contribution from the coupling of orbital angular momentum and the intrinsic spin of the protons and neutrons $s = 1/2$ known as the spin-orbit interaction $l \cdot s$ resulted in a further broken degeneracy which reproduced the observed magic numbers, as seen in the Fig. 2.1.

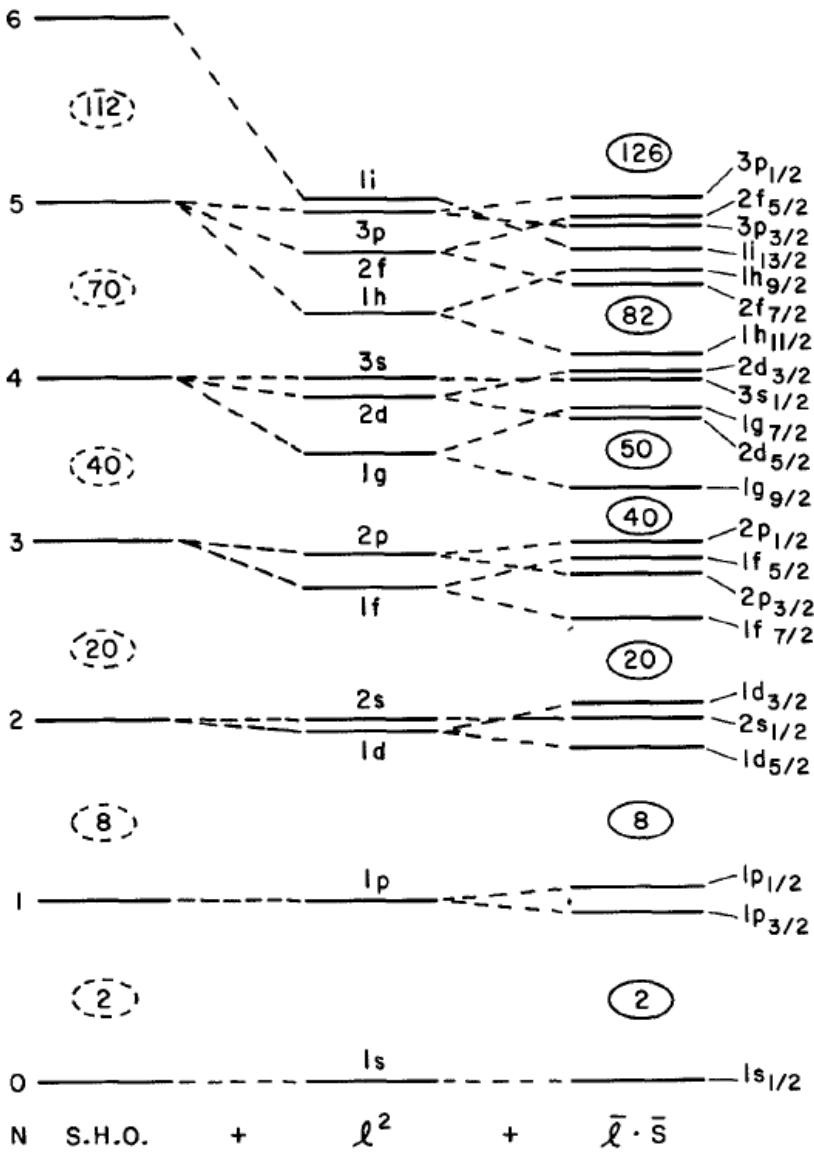


Figure 2.1: Evolution of nuclear shell model orbitals, taken from [28]. See text for more details.

Here, the good quantum numbers are the principal harmonic oscillator number n , orbital angular momentum number l together with the intrinsic spin angular momentum s and the total single-particle angular momentum number $j = l + s$ with degeneracy of $2j + 1$ in each shell model orbit. The total spin of the nucleus I is defined as the vector sum of the single-particle angular momenta j . This shell model provided a theoretical framework for describing microscopic properties of nuclei from the sequential filling of independent proton and neutron shells. The protons or neutrons in each respective orbital energetically favor the formation of spin-zero pairs with opposing spin orientation. This explains why nuclei containing an even number of protons and neutrons systematically have spin-zero ground-states. Consequently, the ground-state spin of nuclei are governed by the orbital(s) occupied by the unpaired nucleon(s). Once a shell model orbit is completely filled, additional energy is required when adding an additional nucleon in a higher-lying orbit, explaining the enhanced binding energy of closed shell nuclei.

While the independent particle shell model is tailored to closed shell nuclei -1 or +1 nucleon, where the residual interaction can largely be neglected, it has been remarkably successful in describing general nuclear properties of isotopes near semi-magic nuclei across the nuclear chart. However, this approximation has its limitations when more nucleons are added beyond the closed shell, where the residual interactions can no longer be ignored.

2.3 The interacting particle shell model and its valence space

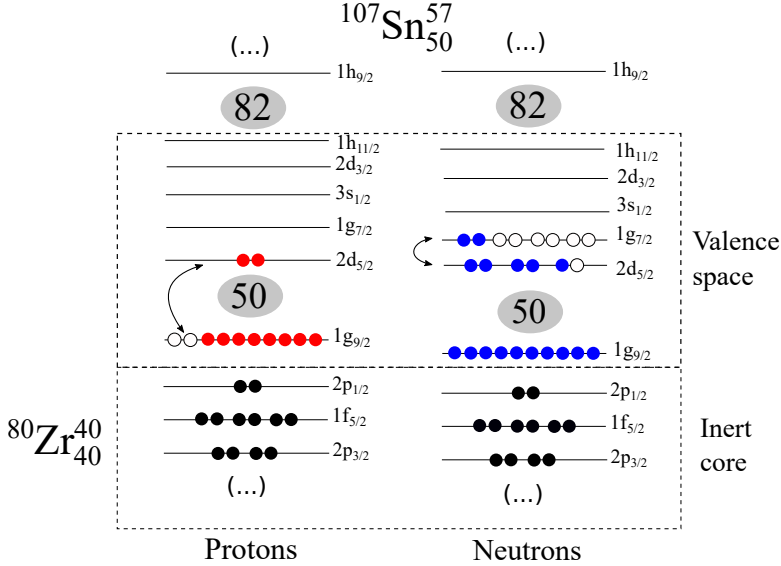


Figure 2.2: Shell model configuration of ^{107}Sn with a selected valence space using a ^{80}Zr core, thus allowing excitations of protons and neutrons from the $1g_{9/2}$ orbit across the $Z = 50$ shell gap.

In principle in a more realistic shell model view, the nucleons outside a doubly-magic core will interact with each other. In this case, the residual nucleon-nucleon interaction has to be considered, to describe the interaction between individual nucleons in a chosen model space. The total wavefunction of the nucleus will in this case be described as a linear combination of single particle wavefunctions σ_i (solutions of the S.E. with the independent particle mean-field Hamiltonian) with probability amplitudes c_i [29],

$$|\Psi(t)\rangle = \sum c_i |\sigma_i\rangle \quad (2.5)$$

The superposition of many states with large coefficients is a signature of configuration mixing, and therefore these type of calculations are also called configuration interaction calculations (similar as in atomic systems for electronic configurations) [30]. Ideally, when these calculations are performed the

interaction of all nucleons should be considered. However, since there is theoretically an infinite number of states this quickly becomes computationally challenging. In order to overcome this challenge, a model space is carefully chosen with an inactive (inert) core. This core is comprised of a nuclear configuration with fully closed shells where all protons and neutrons are 'locked' in place. An appropriate valence space is then chosen where nucleons are free to interact up to a selected upper limit where excitations are unlikely to occur. Doubly-magic nuclei provide ideal cores, since excitations are unlikely to occur beyond the strong shell closures.

This thesis focuses on studying the nuclear properties near one of the least investigated doubly-magic nucleus predicted by the shell model, ^{100}Sn , with the rare property of containing an equal number of protons and neutrons in predicted closed-shell configurations. Normally, this nucleus is chosen as an inert core for a selected valence space which extends up to the next shell closure. However, in order to investigate the influence of excitations beyond the $Z, N = 50$, the valence space can be extended below ^{100}Sn . Fig. 2.2 displays an example of the selected valence space for ^{107}Sn using a ^{80}Zr core, indicating excitation of proton pairs beyond the $Z = 50$ shell closure.

The number of nucleons n occupying the same j orbital can be given by a j^n configuration. With the assumption that the number of nucleon pairs are conserved means that j^n can be reduced to a j^ν -configuration where the so-called seniority number ν describes the number of unpaired nucleons in an orbital state j . The concept of seniority for describing properties of n -body systems was initially developed by Racah [31] for treating electrons in atoms and later generalized in nuclei for multiple degenerate levels by Arima and Ichimura [32]. This seniority scheme have had tremendous success in describing the general trends of nuclear observables from the shell occupancy n and seniority number ν . This is discussed in greater detail in the references [28, 33].

While the restricted valence space significantly reduces the complexity of the eigenvalue problem, the interactive particle shell model calculations remain difficult to solve. In order to construct the interacting particle Hamiltonian, the interaction potential between the individual nucleons needs to be well understood. Most commonly phenomenological potentials are used which are derived from experimental NN scattering data, such as the ones described in the references [34, 35, 36].

A variety of computational techniques have been developed to find a solution to the S.E. by performing a diagonalization of the derived Hamiltonian, such as KSHELL (for details see [37, 38, 29]). However, in recent decades interaction potentials founded on the fundamental principles from QCD have emerged, presented in the following section.

2.4 *Ab-initio* description of the nucleus

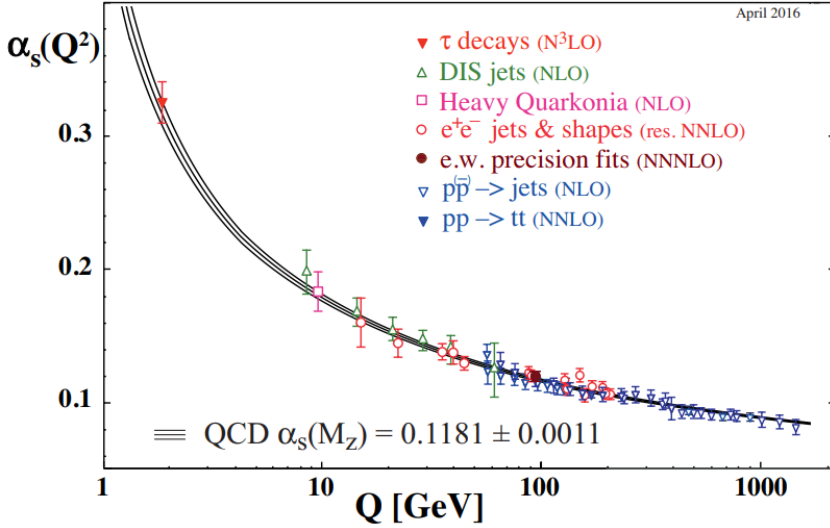


Figure 2.3: The interaction strength of the strong nuclear force α_s with respect to scale energy Q indicating the lacking knowledge of low energy QCD, the scale energy for describing the nucleus is typically < 1 GeV. Diagram taken from [39].

Ideally, the Hamiltonian describing the nucleus should be derived from the fundamental interaction between its nucleons constituents, the quarks and gluons, thus based on the strong, weak and electromagnetic forces as described in the Standard Model [40]. This direct treatment has not yet been realized due to our lacking knowledge of the interactions between gluons and quarks at low energies [41, 42, 43]. While a perturbative treatment of QCD can be applied at high energies (or analogously shorter distances such as the length scale of individual quarks $\sim 10^{-18}$ m), this is not possible at the length scales corresponding to the distances between protons and neutrons ($\sim 10^{-15}$ m) in the nucleus where the interaction strength increases rapidly², see Fig. 2.3. A consistent description of QCD at all energy scales remains one of the great challenges in theoretical nuclear and particle physics.

²The range of a virtual particle is related to its mass and lifetime accordingly, $L \approx c\Delta t \approx \frac{\hbar}{2mc}$, thus the greater the mass (or equivalently energy $E = mc^2$) the shorter the range L , vice verse.

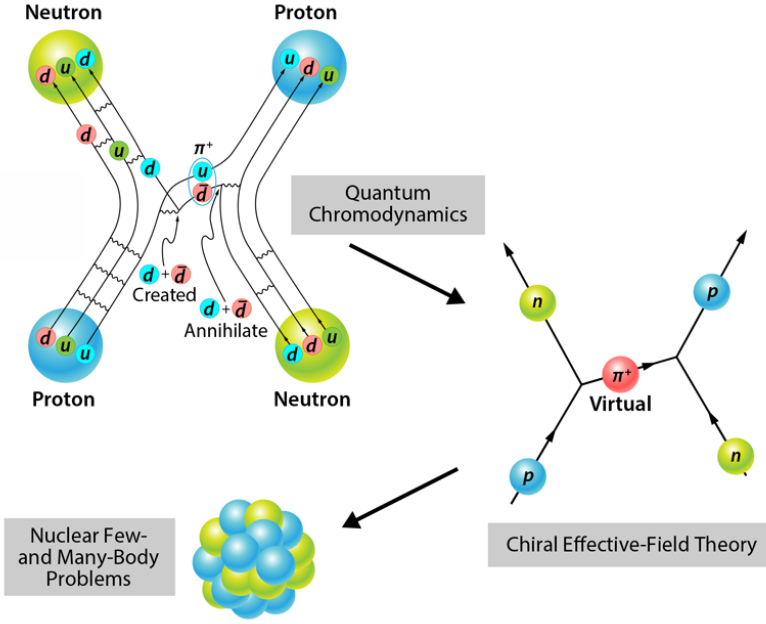


Figure 2.4: Ideal two-step procedure for linking fundamental interactions between quarks (u,d) and gluons to the nuclear many-body problem via χ EFT, using π -meson degrees of freedom. Figure taken from [44].

The quarks and gluons are therefore confined within hadronic matter, such as protons and neutrons inside the nucleus. This means that the quark-gluon interactions can, to a good approximation, be treated as point-like on the scale relevant to describing the properties of the nucleus. With this assumption an effective theory was developed which uses the effective degrees of freedom, such as π -mesons (pions) and nucleons, known as chiral effective field theory (χ EFT), as illustrated in Fig. 2.4(b). With this approach the chiral effective Lagrangian³ is expressed in terms of the interactions between the nucleons and pions,

$$\mathcal{L}_{eff} = \mathcal{L}_{\pi\pi} + \mathcal{L}_{\pi N} + \mathcal{L}_{NN} + \dots \quad (2.6)$$

Where $\pi\pi$ represents pion-pion interactions, and πN describes the interactions between the pion and nucleon etc. Each term is represented by an infinite series of higher order interactions. In order to make sure that the χ EFT Lagrangian describing the nucleus is firmly grounded in QCD, all relevant symmetries

³By convention $\mathcal{L} = T - V$.

and breaking of chiral symmetries are maintained from the underlying QCD Lagrangian (this procedure is described in Ref. [25, 45]). Chiral perturbation theory is used in order to arrange the relevance of the higher order contributions in terms of the scale parameter $(\frac{Q}{\Lambda_\chi})^\nu$ with order ν , where the scale resolution of the effective Lagrangian is given by the ratio of the masses of the exchange particles used as degrees of freedom. The lighter virtual pions correspond to the long-range part of the interaction, often named the soft scale where $Q \approx m_\pi \approx 140$ MeV, and the shorter range of the heavier exchange particles such as ρ -meson defines the scale of the short-range contribution known as the hard scale given by $\Lambda_\chi \approx m_\rho \approx 1$ GeV. In this power counting scheme, represented by Feynman diagrams of increased order as seen in Fig. 2.5, N-body interactions and their relevance naturally emerges in order of the scale parameter with the power ν . This allows for a rough estimate of the missing contributions by calculating the relevance of the higher order diagrams $\nu + 1$. The leading order (LO) $\nu = 0$ is represented by two Feynman diagrams including a cross-vertex from direct interaction of two nucleons and one-pion exchange interaction. The next-leading-order (NLO)⁴ $\nu = 2$ includes two-pion exchange interactions and higher order NN Feynman diagrams. The different vertices in Feynman diagrams are proportional to low energy constants (LEC) of χ EFT which contain the unresolved physics hidden beyond the selected scale and degrees of freedom in the effective theory. The complexity of the interactions as well as the number of LEC increases with increasing order ν . So far, it is not possible to calculate these LEC from a QCD based framework alone. Normally, NN-scattering and ^2H binding energy are used to fix the the LEC required for NN interactions and the binding energy of ^3H and (n, ^2H)-scattering data are used for constraining 3N interactions. Further details can be found in the references [41, 45].

⁴ $\nu = 1$ disappear when parity is conserved [45].

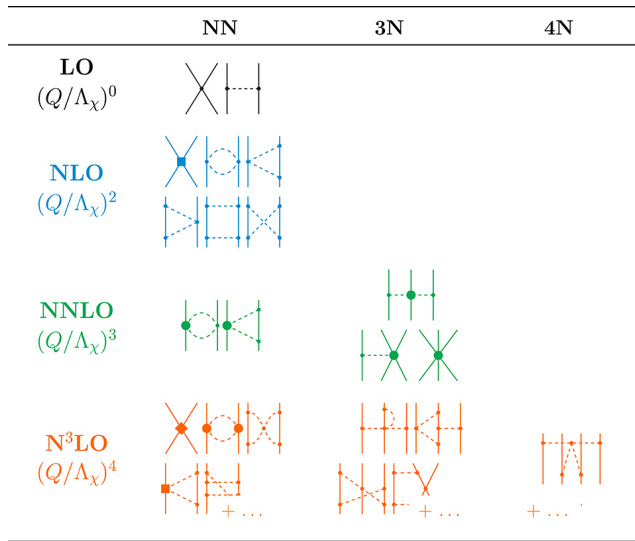


Figure 2.5: Hierarchy of Feynman diagrams where solid lines represent nucleons and dotted represent pions. The small dots, squares, large dots and diamonds represents vertices with increasing index respectively. Taken from Ref. [46].

2.4.1 χ EFT based interactions

Many χ EFT based interactions have been developed over the years and have been successfully employed in different many-body methods for describing nuclear structure properties [46, 47]. In this work two such state-of-the-art *ab-initio* interactions are employed, namely EM1.8/2.0 and Δ NNLOgo described below:

EM1.8/2.0

The EM1.8/2.0 interaction developed by Entem and Machleidt, described in the references [48, 49, 50]. This interaction includes contributions up to N3LO for NN and NNLO for 3N interactions for pions and nucleon degrees of freedom. A 1.8 fm^{-1} momentum cutoff set the resolution for the NN interactions while a 2.0 fm^{-1} momentum cutoff is used for the 3N interactions. The LEC for this interaction have been fitted to reproduce the NN scattering phase shifts, deuteron (^2H) properties, binding energy of 3N systems (^3H , ^3He), as well as the charge radius of ^4He . This interaction have been particularly successful when

employed with many-body calculations methods for calculating nuclear binding energies up to the tin region [27].

Δ NNLOgo

The Δ NNLOgo (or simplified N2LOgo) interaction was developed by the Göteborg-Oakridge collaboration [47]. This interaction is restricted to NNLO for NN and 3N interactions but as the name suggest also includes the heavier $\Delta(1232)$ -isobar in the Feynman diagrams describing these interactions. The LEC of NN and 3N are fitted simultaneously such that it reproduces the low-energy NN phase shifts, few-body data ($A \leq 4$), and the nuclear matter properties [51]. This interaction has recently been applied with many-body methods to calculate charge radii and binding energies with good agreement for potassium isotopes [52].

2.4.2 Solving the many-body problem

Once the free-space interactions, such as the ones described above, have been determined, the Hamiltonian in the S.E. (Eqn. 2.1) can be constructed for the many-body system in question. The endeavour of solving the many-body problem with *ab-initio* based nuclear interactions is by no means a simple task, as it requires the diagonalisation of a matrix of enormous dimensions. Many calculation methods have been developed over the years capable of solving the many-body problem in different fields of physics, such as self-consistent greens function (SCGF), coupled-cluster (CC), In-Medium Similarity Renormalization Group (IMSRG), no-core shell model (NCSM) etc. summarized in [46, 53]. However, the complexity of the many-body problem of N added nucleons in the system means that only lighter nuclei $\leq {}^{16}\text{O}$ could be calculated [54]. In the last decade new computational methods have been developed which allow the treatment of heavier regions of the nuclear chart using the χ EFT based interactions, owing to the rapid development of computationally efficient techniques with controlled truncation points.

One such method employed in this work is the valence-space In-Medium Similarity Renormalization Group (VS-IMSRG), described in detail in [26, 53]. This approach allows an efficient way of solving the many-body problem by decoupling the selected core, valence space and excluded space from the full Hamiltonian derived from the χ EFT based interactions. An effective Hamiltonian is created from the selected valence space in a harmonic oscillator basis. In order to further reduce computational cost the NN and 3N interactions are restricted within a limit of the harmonic oscillator (HO) shells, $e = 2n + l \leq$

e_{max} where the number for the 3N interaction matrix elements are within a boundary limit $e_1 + e_2 + e_3 \leq E_{3max}$. This 3N boundary limit is pushed continuously as it has an increasing importance for improving the accuracy of calculations of heavier nuclei, with the current state-of-the-art reaching $E_{3max}=24$ [55]. Once the unitary transformation of the effective Hamiltonian is available it can be directly applied to any consistent valence space operator for extracting relevant nuclear observable derived using IMSRG++ code [56]. This makes it possible to attack the larger regions of the nuclear chart with an *ab-initio* approach where the effective Hamiltonian is diagonalized using shell model calculations such as KSHELL [38].

Owing to the development of *ab-initio* many-body methods such as VS-IMSRG using interactions derived from χ EFT new regions in the nuclear chart are now accessible. With the current state-of-the-art extending to the tin region and beyond [27], as highlighted in Fig. 2.6. ^{100}Sn is a particularly valuable truncation point for describing the greater regions of the nuclear chart. In order to benchmark these *ab-initio* calculations, experimental data in the proximity of the ^{100}Sn is essential.

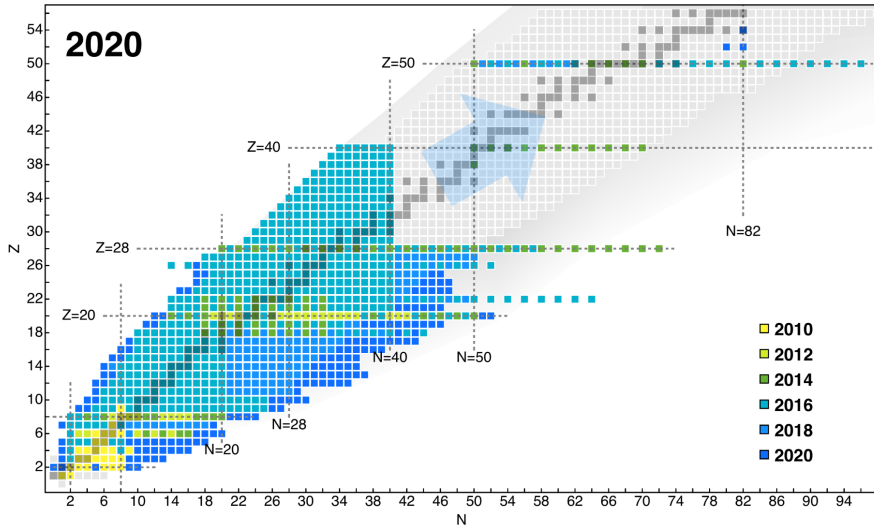


Figure 2.6: Progress of *ab-initio* based many-body calculation in the last decade. Taken from Ref. [46].

2.5 Electromagnetic properties of the nucleus

While the weak and strong nuclear forces are confined within the range of the nucleus, the electromagnetic force extends indefinitely. The motion and distribution of charges within the nucleus are directly linked to the interplay of the weak and strong forces. This means that measurements of the electromagnetic properties are able to reveal the effects of these 'hidden' forces within the nucleus. The electromagnetic properties defined from its charge distribution and currents can be described as a sum of N-body charge ($\rho_e(r)$) and current ($j(r)$) operators,

$$\rho_e(r) = \sum_i \rho_i(r) + \sum_{i<j} \rho_{i,j}(r) + \sum_{i<j<k} \rho_{i,j,k}(r) + \dots \quad (2.7)$$

$$j(r) = \sum_i j_i(r) + \sum_{i<j} j_{i,j}(r) + \sum_{i<j<k} j_{i,j,k}(r) + \dots \quad (2.8)$$

Analogously to the interaction potentials, the one-body operators are well understood while the knowledge of the higher-order contributions are lacking. The complicated charge and current distributions can be simplified by expansion of multipole electric $\hat{Q}_{lm}$ and magnetic $\hat{M}_{lm}$ moments,

$$\hat{Q}_{lm} = \int r_i^l Y_{lm}^*(\theta_i, \omega_i) \rho_e(r) dr \quad (2.9)$$

$$\hat{M}_{lm} = \frac{-1}{l(l+1)} \int j(r) \cdot (r \times \nabla_i) r_i^l Y_{lm}^*(\theta_i, \omega_i) dr \quad (2.10)$$

Where $Y_{lm}^*(\theta_i, \omega_i)$ are the spherical harmonics defining the angular distribution in spherical coordinates with $l = -m, \dots, m$ and r as the radius. Since only multipole terms with even-parity survive the integration over all space, only electric moments with even l and magnetic moments with odd l survive when parity is conserved. The expectation values of the magnetic dipole moment operator $\hat{M}_1$ (or M1) and the electric quadrupole moment operator $\hat{Q}_2$ (or E2) are presented in the following sections.

2.5.1 The magnetic dipole moment and the nuclear g-factor

The magnetic dipole moment is defined as;

$$\mu_I = \langle I, m = I | \hat{M}_1 | I, m = I \rangle = \sum_i^A \left(g_i^{(s)} s_i + g_i^{(l)} l_i \right) \quad (2.11)$$

Where $g_i^{(s)}, g_i^{(l)}$ are the intrinsic and orbital gyromagnetic ratios given by $g_s^p = 5.5856946893(16)$ and $g_l^p = 1$ for a free proton and $g_s^n = -3.82608545(90)$ $g_l^n = 0$ for free neutron respectively [57]. The magnetic dipole moment is given in units of the nuclear magneton $\mu_N = \frac{e\hbar}{2m}$ throughout this thesis work. Furthermore, the magnetic moment can also be given in terms of the g-factor and nuclear spin I as, $\mu = gI$. Measurements of the nuclear magnetic dipole moment (or g-factors) are sensitive to the intrinsic and orbital currents governed by the wavefunctions of the unpaired nucleons in the system. While spin-0 systems exhibit symmetries which cancel external fields, systems with $I > 0$ allow the probing of g-factors. The g-factor assuming a free orbiting proton or neutron may be calculated from the following expression:

$$g_{s,p} = \begin{cases} (j - \frac{1}{2})g_l + \frac{g_s}{2} & \text{for } j = l + 1/2 \\ \frac{j}{j+1} \left[(j + \frac{3}{2})g_l - \frac{g_s}{2} \right] & \text{for } j = l - 1/2 \end{cases} \quad (2.12)$$

These expressions often provide good upper or lower bounds for the g-factor of a nucleon in a given shell model orbital. A lot of information is missed with this naive approximation as the magnetic moment is influenced by the other nucleons which polarize the core as well as missing meson exchange currents which tend to screen the free particle g-factor [58]. Often a quenching factor is used in order to account for these missing effects.

2.5.2 The electric quadrupole moment

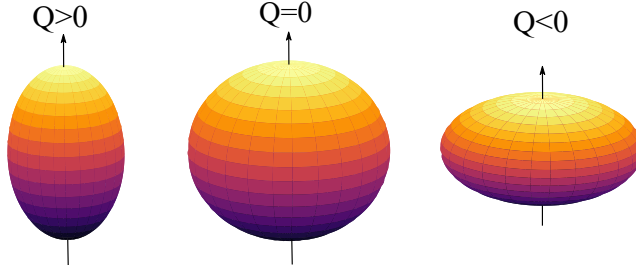


Figure 2.7: The shape of the charge distribution for different electric quadrupole moments.

The spectroscopic quadrupole moment of a nucleus is given by the expectation value of the electric quadrupole operator in the spin aligned state I ,

$$Q_s = \langle I, m = I | \hat{Q}_2 | I, m = I \rangle = \sqrt{\frac{I(2I-1)}{(2I-1)(2I+3)(I+1)}} \langle I || \hat{Q}_z || I \rangle \quad (2.13)$$

Where $\langle I || \hat{Q}_z || I \rangle$ is the reduced matrix element from the Wigner-Eckart theorem. This describes the quadrupole charge deformation of the nucleus which is prolate for positive values and oblate for negative values and spherical when zero, as seen in Fig. 2.7. For a rotating deformed nucleus the time-averaged shape can be distorted in the reference frame of the observer when the rotational axis is different to the nuclear symmetry axis. In order to restore the quadrupole deformation in the intrinsic frame the following transformation can be performed to obtain the intrinsic quadrupole moment Q_0 [58],

$$Q_0 = \frac{(I+1)(2I+3)}{3K^2 - I(I+1)} Q_s \quad (2.14)$$

Where K is the spin projection on the nuclear symmetry axis.

The spectroscopic quadrupole moment resulting from a single nucleon orbiting within the nucleus can be approximated accordingly,

$$Q_{s,s.p} = -\frac{2j-1}{2j+2} \langle r_j^2 \rangle e^{eff}, \quad (2.15)$$

where j is the orbital angular momentum quantum number, $\langle r_j^2 \rangle$ is the mean-square radius of the orbit and e^{eff} is the effective charge⁵ of the orbiting nucleon.

⁵While the neutron carries zero charge it can exhibit an effective charge within the nucleus as it affects the charge distribution of the ensemble.

In this simple interpretation the orbiting nucleon or hole with effective charge e^{eff} polarizes the core of the nucleus, resulting in a quadrupole moment.

This single particle model can be further extended with the seniority scheme for the filling of n particles in an orbit with angular momentum j accordingly [58],

$$Q_s = Q_{s,s,p} \frac{2j+1-2n}{2j+1-2v}, \quad (2.16)$$

For a seniority-1 ($\nu = 1$) configuration a trend of linearly increasing quadrupole moments is expected with added nucleons n in the shell model orbit, with a crossover at zero when the valence orbit is half-filled. This expression can be further generalized to a multi- j configuration where $\Omega = \sum_j \frac{2j+1}{2}$ [59],

$$Q_s = Q_{s,s,p} \frac{\Omega - n}{\Omega - \nu}, \quad (2.17)$$

2.5.3 Charge distribution and the nuclear charge radius

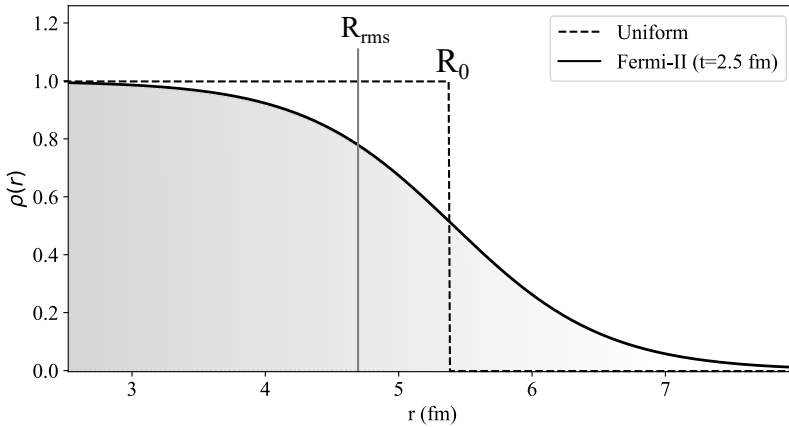


Figure 2.8: The nuclear charge distribution represented by a uniform distribution and the two-parameter Fermi distribution (Eqn. 2.20) with skin thickness t and half-density radius R_0 . The rms charge radius R_{rms} is indicated for the Fermi-II distribution.

One of the most fundamental properties of the nucleus is its size. The size of the nucleus was first determined by Rutherford from scattering of alpha-particles on gold foil [60]. This experiment clarified the approximate size of the nucleus

in the 10^{-15} m length scale. In a spherical drop description of the nucleus the radius of the uniform nuclear sphere is given by,

$$R_0 = r_0 A^{1/3} \text{ fm} \quad (2.18)$$

Where $r_0 = 1.2$ fm provide a good agreement with the general trend in charge radii across the nuclear landscape. However, this simple model quickly breaks down when studying the finer details of isotopic chains.

The size of a nuclear charge distribution can be defined from its radial moments to N^{th} order,

$$\langle r^N \rangle = \frac{\int_0^\infty \rho(\vec{r}) r^N d^3r}{\int_0^\infty \rho(\vec{r}) d^3r}. \quad (2.19)$$

The changes in the second radial moment $N=2$, known as the mean-square charge radius $\langle r^2 \rangle$ is investigated in this work, where the root-mean-square (rms) charge radius is defined as $R_{rms} = \sqrt{\langle r^2 \rangle}$. An accurate description of the charge radius requires complete knowledge of the charge distribution given from the expectation value of the full N-body operator presented in Eqn. 2.5. While this complete description is missing, a simpler approximation is often used which assumes a two-parameter Fermi distribution, identical to the Woods-Saxon potential used in the nuclear shell model [61]:

$$\rho(\vec{r}) = \frac{\rho_0}{1 - e^{\frac{r-R_0}{a}}}. \quad (2.20)$$

Where a is the diffuseness of the charge distribution which can also be given in terms of the skin thickness $t = 4a \ln 3$, and R_0 is the half-density radius. The Fermi distribution with surface thickness t is shown in Fig. 2.8 together with the calculated R_{rms} (a typical skin thickness is on the order of $t = 2.62$ fm for stable tin [62]). The mean-square radius can be approximated from the Fermi distribution as $\langle r^2 \rangle \approx \frac{3}{5} R_0^2 + \frac{7}{5} \pi^2 a^2$ [63]. Since the surface thickness is assumed constant over a range of isotopes the changes in mean-square charge radius can be calculated from $\delta \langle r^2 \rangle \approx \frac{3}{5} \delta R_0^2$.

The charge radius provides essential insights into the nuclear structure, for example shell closures can be clearly identified from the sudden enhanced radius with added nucleons beyond the shell. These phenomena are discussed in the context of tin later in this thesis (Sec. 8.4).

2.6 Dynamic and static deformation of the nucleus

The nucleus can exhibit collective properties such as phonon modes of vibration of its surface. These dynamic deformations of the nuclear surface results in an effective increase in the distribution of charge. Assuming a nucleus described as a spherical drop with radius R_0 the effective increase in its size caused by phonon vibrations can be expressed as [64],

$$R(\theta, \omega) = R_0[1 + \sum_l \sum_{m=-l}^{m=+l} \alpha_{lm}^*(t) Y_{lm}(\theta, \omega)] \quad (2.21)$$

Where the deformation parameter is given by $\alpha_{i0} = \beta_i$. The mean-square charge radius can be decomposed into a spherical and deformed contribution as presented in the general form:

$$\langle r^2 \rangle \approx \langle r_0^2 \rangle \left[1 + \frac{5}{4\pi} \sum_i \langle \beta_i^2 \rangle \right]. \quad (2.22)$$

Where $\langle r_0^2 \rangle$ is the spherical contribution together with the deformation contribution in terms of the β deformation parameters. In most cases only quadrupole vibrations need to be considered since higher orders have a vanishing contribution. Eqn. 2.22 can be reshuffled for extracting deformation if the spherical contribution of the mean-square charge radius is known,

$$\sum_i \langle \beta_i^2 \rangle \approx \frac{4\pi}{5\langle r_0^2 \rangle} (\langle r^2 \rangle - \langle r_0^2 \rangle) \quad (2.23)$$

This provides a useful phenomenological view of how nuclear deformation can affect the mean-square charge radius, and has been successful in describing the deformation in different regions of the nuclear chart such as the Po and Ba isotopic chains [65, 66].

Another approach to investigate dynamic deformation is by studying the probability of excitation to the collective modes of deformation. The transition probability to quadrupole excitations in nuclei from initial state I_i to the final I_f state is defined accordingly [28],

$$B(E2 : I_i \rightarrow I_f) = \frac{1}{2I_i + 1} \langle I_f || \hat{Q}_2 || I_i \rangle^2 \quad (2.24)$$

The reduced quadrupole transition probability from the ground state to the first excited state in even-even nuclei $B(E2 : 0_{gs}^+ \rightarrow 2_1^+)$ gives an indication of the softness to dynamic quadrupole deformation of the nucleus, or similarly its collectivity. This quantity can be directly related to the quadrupole beta-deformation parameter according to the hydrodynamical model assumption of a vibrating sphere [64],

$$\langle \beta_2 \rangle = B(E2 : 0_{gs}^+ \rightarrow 2_1^+)^{1/2} \frac{4\pi}{3ZR_0^2} \quad (2.25)$$

Where R_0 is the equivalent radius of the spherical contribution and Z is the total charge of the nucleus.

Similarly, the quadrupole deformation parameter resulting from the static intrinsic deformation can be determined from measurements of the quadrupole moment, as presented earlier in Eqn. 2.14. The intrinsic deformation can be expressed in terms of the β_2 parameter accordingly [58],

$$Q_0 \approx \frac{3}{\sqrt{5\pi}} Z R_0^2 \beta_2^{static} \quad (2.26)$$

Which results in the expression,

$$\beta_2^{static} \approx \frac{\sqrt{5\pi}}{3ZR_0^2} Q_0 \quad (2.27)$$

While these different phenomenological methods are highly model dependant they provide useful tools for studying the extent of collective nuclear deformations in nuclei [67]. The total deformation of the nucleus is thus given by,

$$(\beta_i^{total})^2 = (\beta_i^{static})^2 + (\beta_i^{dynamic})^2 \quad (2.28)$$

In the case of spin-0 nuclei the static deformation is not measurable from the quadrupole moments and the dynamic contribution is governing the total deformation of the nucleus.

2.7 The interest in the tin isotopic chain

In order to establish ^{100}Sn as a valid truncation point for many-body calculations approaches it is essential to investigate the nuclear structure of its neighbouring nuclei. While multiple measurements have confirmed the doubly-magic character of ^{132}Sn with 50 protons and 82 neutrons [14, 15, 16, 17, 18], there is no conclusive evidence of the robustness of the shell closures of the short-lived ^{100}Sn at the neutron-deficient end of the tin chain. Experimental efforts lasting several decades have striven towards a better understanding of the neutron-deficient tin region. A review of experimental effort is found in Ref. [68]. Measurements of the super-allowed beta-decay of ^{100}Sn were successfully performed by C. Hinke *et al.* [8] which was more recently remeasured by D. Lubos *et al.* [69]. These measurements together with recent mass measurements of ^{100}In , provide constraints of the mass of ^{100}Sn [7]. These results are of particularly valuable for modelling astrophysical scenarios. Since ^{100}Sn is predicted as the end of the rapid proton capture process in nucleosynthesis, these measurements provide insight for explaining the abundances of the elements in our universe [68]. No other properties of ^{100}Sn have been published so far, leaving many questions unanswered. Two of those are of particular interest in this thesis:

Neutron level ordering

The arrangement of the closely spaced $2d_{5/2}$ and $1g_{7/2}$ neutron orbitals above $N=50$ in Sn are currently unknown, see shell model scheme in Fig. 2.9. Understanding the evolution of the ordering of these two levels above ^{100}Sn is crucial for benchmarking many-body calculations and for clarifying the structure of nuclei around ^{100}Sn . Gamma spectroscopy measurements by D. Seweryniak *et al.* [23] suggest a $5/2^+$ ground-state in ^{101}Sn corresponding to an unpaired neutron occupying the $2d_{5/2}$ orbital. However, measurements by I. Darby *et al.* [22] revealed a $7/2^+$ ground-state in ^{101}Sn from subsequent alpha-decays of ^{109}Te , which adds to the confusion. The ground-states of several odd- N tin isotopes, ^{103}Sn [70, 71], ^{105}Sn [72, 73] and ^{107}Sn [74, 75], have only been tentatively assigned ($5/2^+$) from gamma and beta-decay spectroscopy measurements. Furthermore, many-body calculations using CC and VS-IMSRG suggest a different evolution of the ordering of the two orbitals, depending on the interaction used, presented by T.D. Morris *et al.* in Ref. [27]. So far, nuclear model independent ground-state spins have only been firmly established down to ^{109}Sn using laser spectroscopy techniques performed by J. Eberz *et al.* [76] and more recently confirmed by D.T. Yordanov *et al.* [77].

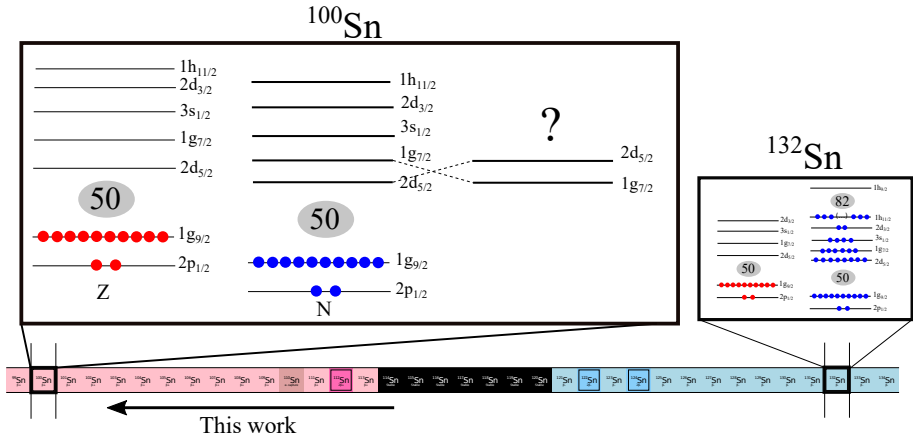


Figure 2.9: The tin isotopic chain highlighting the shell model configuration for ^{100}Sn and ^{132}Sn . The ordering of the lowest lying neutron orbitals above ^{100}Sn are unknown. This work aims to investigate the evolution of ground-state nuclear properties approaching ^{100}Sn .

Collective effects approaching ^{100}Sn

Doubly-magic nuclei have characteristically rigid spherical structures. The added or removed neutrons beyond the closed shells are therefore expected to drive the collective properties of the nucleus. Consequently, a trend of maximized collective effects is expected in between the two major shell closures from the generalized seniority filling of the shell model orbitals between $N=50,82$. The evolution of collectivity along the tin chain has been studied extensively by probing the quadrupole transition probability $B(E2)$ of even-even tin isotopes [21, 78, 79, 19, 20, 80, 81, 82, 83], as seen in Fig. 2.10. While there is a seemingly smooth trend in decreased $B(E2)$ approaching $N=82$ there is a notable deviation on the neutron-deficient side from the parabolic trend predicted by Large scale shell model (LSSM) calculations assuming a ^{100}Sn core [19]. The notable asymmetry indicates an enhanced softness to dynamic quadrupole deformation with reduced neutron number, when approaching ^{100}Sn . Interestingly, while a $B(E2)$ measurement of ^{104}Sn by G. Guastalla *et al.* [79] indicates a clear reduction in quadrupole collectivity, other measurements show only a moderate decrease Ref. [20, 21]. These contradictory measurements further add to the uncertainty of the robustness of the ^{100}Sn shell-closures.

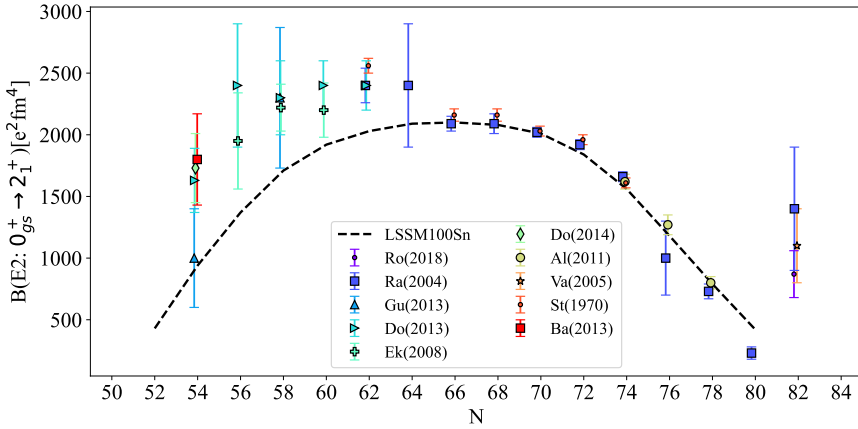


Figure 2.10: Measured $B(E2)$ transition probabilities along the tin isotopic chain using data from [78, 79, 21, 19, 20, 80, 81, 82, 83]. The data is presented together with calculated $B(E2)$ values from Large Scale Shell model calculations assuming a ^{100}Sn core, taken from Ref. [19]. There is a noticeable asymmetry in the parabolic trend where experiments indicate an enhanced $B(E2)$ towards the neutron-deficient tin isotopes.

Summary

Measurements of magnetic dipole moments and spins are particularly sensitive to the orbital of the unpaired neutrons and can therefore provide a particularly useful insight into the ground-state level ordering of the lightest tin isotopes. Furthermore, the evolution of collective properties can be investigated by probing nuclear charge radii and quadrupole moments which could provide a new perspective on the enhanced softness to deformation approaching ^{100}Sn . Measurements of these electromagnetic properties of the nucleus can be investigated by probing the orbiting electrons using laser spectroscopy, which is described in the following chapter. Nuclear moments and charge radii have been extensively studied in the neutron-rich region extending across $^{109-134}\text{Sn}$ [77, 15, 17, 84, 85, 76]. This work aim to fill the missing gap on the neutron-deficient side of the tin isotopic chain by performing measurements of nuclear moments, spins and charge radii below ^{108}Sn for the first time.

Chapter 3

Atomic electrons and their interactions with the nucleus

Electrons bound in the electrostatic potential of the nucleus can be studied with sufficient precision to reveal nuclear properties such as nuclear electromagnetic moments, mean-square charge radii and spin of particular interest in this thesis. In this chapter, the electromagnetic interaction between the nucleus and its bound electrons in a free atom is described. The insight into the minuscule perturbations of the electronic energy levels caused by nuclear structural changes, reveals effects such as isotope shifts and hyperfine splittings which can be studied using spectroscopic techniques. The procedure used for extracting the nuclear observables from spectroscopic measurements of the hyperfine perturbations of atomic levels are presented here.

3.1 Properties of bound electrons in an atom

The nucleus is roughly $1/100000$ th the size of the surrounding electron cloud of the atom, therefore it is often justified to approximate the nucleus as an infinitesimal point with only two properties, its charge and mass. The electrons in the atom are bound in discrete energy levels given by the electrostatic potential of the charged nucleus. The main contribution to these states is the radial distance from the nucleus which is quantized according to the principal quantum number $n = 1, 2, 3, \dots$. For each principal level the electron can have different orbital angular momenta given by the azimuthal quantum number $l = 0, 1, \dots, n - 1$, commonly labeled $s, p, d, f, g, \dots$ for historical reasons [86]. The

energy spacings between these atomic levels labeled nl are in the order of a few eV and reduces with increased distance from the nucleus. A more detailed sub-structure emerges when taking into account the intrinsic spin of the electron $s = 1/2$ as it couples to the electron orbital angular momentum, known as the spin-orbit interaction. This can be further generalized to the interaction of total orbital ($\mathbf{L} = \sum \mathbf{l}_i$) and spin ($\mathbf{S} = \sum \mathbf{s}_i$) angular momenta from multiple unpaired electrons, coupling to the total angular momentum $\mathbf{J}$, where $|L - S| < J < L + S$ with integer steps. These so called fine structure states are commonly labeled according to the Russell–Saunders coupling scheme $nl^{2S+1}L_J$, where $2S + 1$ is the multiplicity of the state given by the number of unpaired electrons. The fine-structure splittings of the electronic energy levels are typically in the order of 0.1 meV for light systems and increase up to 100s of meV for heavier many-electron systems [87].

In the following section the consequence of having a nucleus with finite size and properties beyond its charge and mass is explored, such as its charge radius, spin and electromagnetic moments. The hyperfine perturbations of the atomic energy levels caused by these nuclear properties are about 1000 times smaller than the atomic fine structure splittings, however, these perturbations when probed, allow nuclear-model independent insights into the nuclear properties, such as the tin isotopes of interest in this work.

3.2 Electron-nucleus interactions

The Hamiltonian operator describing the total energy of an atom can be separated into the Hamiltonian of a free nucleus $\hat{H}_n$, the electron cloud $\hat{H}_e$ and the interactions between the nucleus and the electrons $\hat{H}_{ne}$ which will be explored further. While the interactions between nucleons are a complex interplay between the electromagnetic, strong and weak nuclear force, the interaction between the electrons and the nucleus is assumed to be purely electromagnetic¹. The electromagnetic interaction Hamiltonian can be simplified by separating the electrostatic ($\hat{H}_{cc}$ charge-charge) and magneto-static ($\hat{H}_{jj}$ current-current) interactions. If the exact charge distribution of the nucleus and the electrons are known then the charge-charge interaction Hamiltonian $\hat{H}_{cc}$ can be determined, but this is in principle not the case. Instead, the charge distributions are expanded as a series of rank n electrostatic multipole tensors, separating the contribution from the electron cloud $\hat{V}^{(n)}$ (electric field tensors) and the nuclear charge distribution operator $\hat{Q}^{(n)}$ (Eqn. 2.5). Likewise, the Hamiltonian of the current-current interaction $\hat{H}_{JJ}$ can be described as a series

¹With sufficient precision the weak interaction may come into play, this interaction is however negligible under the circumstances discussed in this thesis [88, 89].

expansion of the magnetostatic moment and field tensors $\hat{M}^{(n)}$ (Eqn. 2.5) and $\hat{B}^{(n)}$ for the nuclear magnetic moments and magnetic fields generated by the electrons respectively, this procedure is described in greater detail in the References [90, 91, 92]. The nuclear-electron interaction Hamiltonian can thus be presented as the infinite series expansion of higher order moments:

$$\hat{H}_{ne} = \underbrace{\sum_{n=0}^{\infty} \frac{\hat{Q}^{(n)} \otimes \hat{V}^{(n)}}{2n+1}}_{\hat{H}_{qq}} + \underbrace{\sum_{n=0}^{\infty} \frac{\hat{B}^{(n)} \cdot \hat{M}^{(n)}}{2n+1}}_{\hat{H}_{jj}}. \quad (3.1)$$

Here, the charge and current interactions are expanded in multipole series of tensor and scalar products of the nuclear and electronic moments. Since only multipole terms with even-parity survive the integration over all space when calculating the expectation value, only terms with even- n terms remain for the charge-charge interactions and odd- n for the current-current interaction, thus no magnetic monopole term or electric dipole moment exist when parity is conserved. Therefore, the remaining operators describing the electron-nucleus Hamiltonian are provided in Eqn. 3.2 where the higher order contributions have rapidly reducing influence to the energy levels.

$$\hat{H}_{ne} \approx \underbrace{\hat{Q}^{(0)} \otimes \hat{V}^{(0)}}_{\hat{H}_{E0}} + \underbrace{\hat{B}^{(1)} \cdot \hat{M}^{(1)}}_{\hat{H}_{M1}} + \underbrace{\hat{Q}^{(2)} \otimes \hat{V}^{(2)}}_{\hat{H}_{E2}} + \dots \quad (3.2)$$

Where the $\hat{H}_{E0}$ is the electric monopole, $\hat{H}_{M1}$ is the magnetic dipole and $\hat{H}_{E2}$ is the electric quadrupole moment. While the monopole term is dominating and leading to a shift of the atomic energy levels, the higher order multipole moments can be approximated as perturbations of the full Hamiltonian. Perturbation theory is therefore used when calculating the contributions from the higher order moments.

3.3 The monopole moment

The electric monopole term is the dominant term for describing the electronic structure. The expectation value of the electric monopole operator can be approximated assuming a nucleus of finite size with N number of electrons and Z number of protons [90]:

$$E_{(E0)} = \langle \hat{H}_{E0} \rangle \approx \underbrace{-\frac{e^2 N Z}{4\pi\epsilon_0} \left\langle \frac{1}{r_e} \right\rangle}_{r_e > r} - \underbrace{\frac{eZ}{6\epsilon_0} \rho(\vec{0}) \langle r^2 \rangle}_{r_e < r}. \quad (3.3)$$

Here, the first term corresponds to the electrostatic potential of the electron cloud at the average radius r_e away from the nucleus described as a point charged nucleus, the second term represents the first order electric monopole correction for when the electronic wavefunction is overlapping within a finite sized nucleus with mean-square charge radius². The average electron charge density at the nucleus $\rho(\vec{0}) = Ze|\psi_e(\vec{0})|^2$ varies depending on the electronic wavefunction. In the non-relativistic case only the s-electrons have a finite overlap with the nucleus, while in relativistic systems p-electrons also have an overlapping distribution within the nucleus. This overlapping charge distribution contribution called the first order monopole shift (Eqn. 3.3), effectively reduces the Coulomb attraction experienced by the electrons orbiting the nucleus, resulting in a shift of the energy level.

3.3.1 The field-shift

While the absolute shift of an atomic energy level is difficult to measure directly, measurements of transition energies ν_0 between two atomic states ψ_i and ψ_f are regularly performed using spectroscopic techniques described in Sec. 3.5. Eqn. 3.3 indicates that the second term is sensitive to the the mean-square charge radius of the nucleus, thus small changes in the nuclear charge radius $\delta\langle r^2 \rangle = \langle r^2 \rangle^{A'} - \langle r^2 \rangle^A$ between two nuclei A, A' , will result in a change in transition frequency $\delta\nu_{FS}$ known as the field-shift (FS) effect. The expression for the FS measured between the initial and final atomic states is given by,

$$\delta\nu_{FS} = -\frac{e^2 Z}{6\epsilon_0} \delta|\psi_e(\vec{0})|_{i \rightarrow f}^2 \delta\langle r^2 \rangle^{A, A'} = F_\lambda \delta\langle r^2 \rangle^{A, A'}. \quad (3.4)$$

Here, the contribution from the electrons and the nucleus are separated in the electronic FS factor F_λ , which is assumed constant with respect to an atomic transition in an element, and the changes in the mean-square nuclear charge radius $\delta\langle r^2 \rangle^{A, A'}$. The FS factor indicates the sensitivity of a transition to the change in size of the nuclear charge distribution. This expression only takes the first order monopole shift into account, here the electron density at the nucleus is assumed constant. A term accounting for the relativistic corrections is added when describing heavier atomic systems. This approximation breaks down when the overlapping charge volume is significant³. Seltzer [94], described the overlapping electric charge distribution in terms of a polynomial expansion of radial moments (Eqn. 2.5.3) for $N = 2, 4, 6, \dots$ with coefficients known as

²Higher-order multipole corrections, describing the shape of the overlapping charge distributions are neglected here but enter as corrections in 3.5.

³This is the case for muonic atoms where the muon charge distribution has a considerable overlap with the nuclear volume due to the increased muon mass compared to electrons [93].

the Seltzer coefficients C_n , to account for the shape of the overlapping charge distribution within the nuclear volume. These contributions can be added as corrections to the expression for the FS factor [93]:

$$\delta\nu_{FS}^{A,A'} = F_\lambda \delta\langle r^2 \rangle^{A,A'} \left(1 + \frac{C_2}{C_1} \frac{\delta\langle r^4 \rangle^{A,A'}}{\delta\langle r^2 \rangle^{A,A'}} + \frac{C_3}{C_1} \frac{\delta\langle r^6 \rangle^{A,A'}}{\delta\langle r^2 \rangle^{A,A'}} + \dots \right) \quad (3.5)$$

Where the coefficients C_n can be accurately calculated, as described by Fricke *et al.* [93]. Generally, these terms are neglected for light to medium heavy nuclei. In tin they account for a correction of 2.7% to the mean-square charge radius [62].

3.3.2 The mass and isotope shift

The change in mass of the atomic system when adding or removing neutrons from the nucleus results in a mass-shift to the fine structure levels. There are two dominant contributions to the mass-shift (MS) effect, namely the normal MS and the specific MS. The normal MS is calculated from the one-body kinetic energy operator accordingly $\frac{m_{A'} - m_A}{m_{A'} m_A} \frac{1}{2} \langle \sum_i p_i^2 \rangle$, which describes the change in kinetic energy of the electronic orbital resulting from the change in reduced mass of the atomic system. This term is large for lighter mass nuclei where the fractional mass of an additional neutron contributes significantly to the change in mass of the atom, such as hydrogen where the normal MS is even greater than the separation between fine-structure levels.

The specific MS is an effect occurring in multi-electron atoms and is represented by the two-body operator $\frac{m_{A'} - m_A}{m_{A'} m_A} \langle \sum_{i>j} p_i \cdot p_j \rangle$, which describes how the kinetic energy of the highly correlated motion of the electrons changes with the change in reduced mass. The specific MS in many electron systems represent a large scale many-body problem and is notoriously challenging to calculate. The non-relativistic total MS is given by the sum of the two contributions:

$$\delta\nu_{MS}^{A,A'} = \mu^{A,A'} \left(\frac{1}{2} \langle \sum p_i^2 \rangle + \langle \sum p_i \cdot p_j \rangle \right) = \mu^{A,A'} \underbrace{(K_{NMS} + K_{SMS})}_{K_{MS}}. \quad (3.6)$$

Where $\mu^{A,A'}$ is the reduced mass factor given by,

$$\mu^{A,A'} = \frac{m_{A'} - m_A}{m_{A'} m_A}. \quad (3.7)$$

The combined electronic mass-shift $k_{MS} = k_{NMS} + k_{SMS}$ is constant for an element when adding or removing neutrons from the nucleus.

The experimentally measurable change in fine structure frequency between two isotopes A, A' of the same element is the result from the combined contribution of the FS and MS effects, named the isotope shift (IS):

$$\delta\nu_{IS}^{A,A'} = \delta\nu_{FS}^{A,A'} + \delta\nu_{MS}^{A,A'} \quad (3.8)$$

This expression can be expanded using Eqn. 3.4 and 3.6 resulting in the commonly used approximation ignoring the higher-order moment corrections,

$$\delta\nu_{IS}^{A,A'} = F_\lambda \delta\langle r^2 \rangle^{A,A'} + k_{MS} \mu^{A,A'}. \quad (3.9)$$

This equation describes the change of the atomic transition frequency between two isotopes A, A' in terms of the electronic factors F_λ and K_{MS} and the nuclear observables $\delta\langle r^2 \rangle^{A,A'}$ and $m_{A'}, m_A$. This expression is used for probing the mean-square charge radius in this work, described further in Sec. 3.5.1.

3.4 The hyperfine structure

While the electric monopole term contributes only to a shift of the fine structure atomic levels as described in the previously, perturbations caused by the higher-order multipole terms,

$$\hat{H}_{HFS} = \hat{H}_{M1} + \hat{H}_{E2} + \dots \quad (3.10)$$

contribute to breaking the degeneracy of the atomic fine-structure levels into a substructure called the hyperfine structure. The additional coupling of the total electron angular momentum J with the nuclear spin I form hyperfine structure levels of the total angular momentum quantum number F given in integers steps from $|J - I| < F < I + J$. The hyperfine structure is dominated by the lower order terms in the multipole expansion. Only two of these effects are measurable with the spectroscopic technique discussed in this thesis, namely the magnetic dipole contribution $\hat{H}_{M1}$ and the electric quadrupole interaction $\hat{H}_{E2}$ which will be developed further, while any higher contribution are neglected in this work.

3.4.1 The magnetic dipole interaction

For an atomic system with $I, J > 0$, the energy splittings of the fine-structure levels resulting from the magnetic dipole interaction between the electrons and nucleus can be expressed with the Hamiltonian,

$$\hat{H}_{M1} = - \int \mathbf{B}_e \cdot d\boldsymbol{\mu}_I \quad (3.11)$$

Here $\mathbf{B}_e$ represent the magnetic field generated by the electrons and $\boldsymbol{\mu}_I$ the nuclear magnetic moment vector. Assuming a constant magnetic field over the nuclear volume and uniform magnetisation of the nucleus, the expectation value of Eqn. 3.11 for an electronic level with total angular momentum F , nuclear spin I and electronic spin J can be expressed as follows,

$$\Delta E_{M1} = \langle \hat{H}_{M1} \rangle = -\frac{1}{2} A_{hf} C \quad (3.12)$$

$$C = F(F+1) - J(J+1) - I(I+1). \quad (3.13)$$

Where C indicates the orientation of the nuclear spin I with respect to the electron angular momentum J . The hyperfine parameter A_{hf} is expressed accordingly,

$$A_{hf} = \frac{\mu_I B_0}{IJ} = g A_0 \quad (3.14)$$

This parameter can be factorized into the nuclear g-factor $g = \mu_I/I$ and the electronic contribution $A_0 = B_0/J$, where B_0 is the average magnetic field at the nucleus generated by the electrons. The approximation assumes a point magnetic dipole with uniform magnetisation over the nuclear volume (B_e assumed constant over the nuclear volume, where it is set to B_0). Further corrections are added to account for the non-uniform magnetization over the nuclear volume (if B_e is not constant, it cannot be placed in front of the integral), known as the Bohr-Weisskopf effect [95] and the modification from the extended nuclear charge distribution known as the Breit-Rosenthal-Crawford-Schawlow correction [96]. These contributions can be added as corrections to the hyperfine parameter accordingly,

$$A_{hf} = A_{hf}^{(point)} (1 + \epsilon_{BW}) (1 + \epsilon_{BR}) \quad (3.15)$$

In the case of tin these contributions are expected to be negligible compared to the point approximation, discussed further in Sec. 7.3.1.

3.4.2 The electric quadrupole interaction

When $I, J > 1/2$ the electric field gradient generated by the charge distribution of the electrons interacts with the electric quadrupole moment of the nucleus adding an additional contribution to the hyperfine structure splittings. This contribution can be expressed from the expectation value of the electric quadrupole Hamiltonian;

$$\Delta E_{E2} = \langle \hat{H}_{E2} \rangle = B_{hf} \frac{3(C(C+1) - 4I(I+1)J(J+1))}{8IJ(2I-1)(2J-1)}, \quad (3.16)$$

$$B_{hf} = eQ_s V_{zz} = qQ_s. \quad (3.17)$$

Where Q_s is the spectroscopic quadrupole moment of the nucleus and V_{zz} (sometimes given in terms of q) is the electric field gradient at the location of the nucleus. This electric field gradient is assumed constant over the nuclear volume which is a good approximation for electronic systems with the precision of the techniques discussed in this thesis.

3.4.3 Higher-order multipole moments

While only the first allowed multipole terms (E0,M1,E2) provide observables relevant for the technique discussed in this thesis, there are infinitely many smaller multipole moments with subsequently reduced energy contribution with increasing order. However, with sufficient resolution the next allowed multipole contribution, the magnetic octupole moment, can be measured in atomic systems where $I, J > 1$. This has only been performed in few atomic systems and requires specialized spectroscopic techniques [97, 98, 99]. These measurements provide an additional interesting insight into the nuclear structure and the fundamental forces involved [99]. So far, only the electromagnetic force has been taken into account, which strictly conserves parity symmetry. However, if the weak interaction is considered as well, the parity (P) and charge-parity (CP) are no longer conserved and the odd-moments (M0,E1,M2,E3...) could hypothetically be observed [100, 101, 102]. The search for these symmetry-violating moments is at the forefront of physics for bench-marking our understanding of the standard model and could answer major questions in physics such as the broken matter-antimatter symmetry in our universe. Ultra-high precision measurements of atoms and molecules show promising developments which could offer sufficient sensitivities for probing these quantities in the coming future [103].

3.5 Extracting nuclear observables using laser spectroscopy

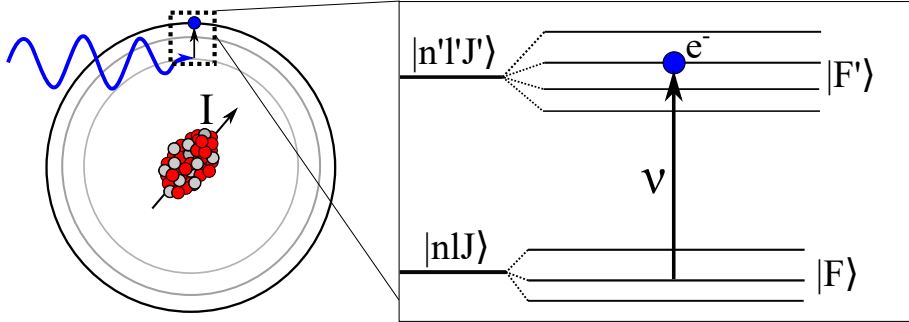


Figure 3.1: The electronic fine structure J levels are split into hyperfine states F from the coupling with the nuclear spin I . Spectroscopy involves excitation among the hyperfine F states.

Measurements of IS and hyperfine structures can provide insight into nuclear structure observables such as changes in mean-square charge radii, magnetic dipole moments, electric quadrupole moments as well as nuclear spin. Despite the small influence of only 10^{-6} eV to the atomic levels these features have been studied for over 140 years at the time of writing [104]. The development of lasers over half a century ago provided a monochromatic light source with tunable frequency which allow scanning across the transition frequency between fine-structure levels $n l J \rightarrow n' l' J'$ which could reveal transitions between hyperfine sub-states $F \rightarrow F'$ when $I, J > 0$, see Fig. 3.1. The probability for exciting an electron configuration from a fine structure level F (in electron state J) into another hyperfine level F' (in electronic level J') can be measured from the subsequent fluorescence decay (emitting optical photons) or by counting ions after (resonance) ionization of the excited level. Thus, the hyperfine structure transition are revealed, as described in Sec. 4.1. This section describes how the nuclear observables I, Q_s, μ_I and $\delta\langle r^2 \rangle$ can be extracted from laser spectroscopic measurements of the isotope shift and the hyperfine structure.

3.5.1 Nuclear charge radii from isotope shift measurements

The IS of the atomic fine structure levels are a result of the change in nuclear charge distribution and mass of the nucleus held within the atom. While accurate measurements of atomic masses have been performed using a variety of sensitive

experimental techniques across the nuclear chart [105], precision measurements of absolute nuclear charge radii from muonic atoms and electron scattering measurements are often limited to stable isotopes [106]. If the electronic factors F_λ , k_{MS} are known for an atomic transition in an element, then the difference in mean-square charge radii along the isotopic chain can be extracted from measurements of IS using Eqn. 3.9 accordingly;

$$\delta\langle r^2 \rangle^{A,A'} = \frac{1}{F_\lambda} (\delta\nu^{A,A'} - \mu^{A,A'} k_{\text{MS}}). \quad (3.18)$$

The FS factor F_λ can currently be calculated with good precision thanks to recent advancements in quantum chemistry methods such as relativistic CC methods [107, 108]. The MS is particularly challenging to calculate with high precision for many electron systems, since the specific MS represents a highly correlated many-body problem. Recent progress using configuration interaction with multi-body perturbation theory has proved successful for calculating the specific MS in medium heavy elements [107, 109]. When the electronic factors are determined for one transition in an element, the factors of the other transition can be deduced by comparing the IS of the same isotopes, provided that IS have been measured for three or more isotopes. Using Eqn. 3.18 for the two atomic transitions (1) and (2) result in the expression:

$$\Lambda_{(2)}^{A,A'} = \frac{F_\lambda^{(2)}}{F_\lambda^{(1)}} \Lambda_{(1)}^{A,A'} + k_{\text{MS}}^{(2)} - \frac{F_\lambda^{(2)}}{F_\lambda^{(1)}} k_{\text{MS}}^{(1)} = \frac{F_\lambda^{(2)}}{F_\lambda^{(1)}} \Lambda_{(1)}^{A,A'} + C \quad (3.19)$$

Where $\Lambda^{A,A'} = \delta\nu^{A,A'} [\mu^{A,A'}]^{-1}$ is defined as the modified IS. The modified IS of the two transitions are linearly dependent with slope $\frac{F_\lambda^{(2)}}{F_\lambda^{(1)}}$ and intersect $C = k_{\text{MS}}^{(2)} - \frac{F_\lambda^{(2)}}{F_\lambda^{(1)}} k_{\text{MS}}^{(1)}$.

Thus, both FS and MS factors can be extracted using linear regression of the modified IS if the electronic factors of one of the transitions are known. This procedure is typically known as the king-plot (KP) method [110]. It should be noted that this procedure is dependant on the accuracy of the electronic factors for one transition.

Elements containing long chains of stable isotopes often have accurately measured absolute mean-square charge radii extracted from muonic or kaonic x-ray spectroscopy and electron scattering techniques. A combined analysis described by Fricke *et al.* in [93] is often used. Here the radial moments V_2^e derived from the charge distribution acquired from electron scattering and the Barrett radii from $R_{\kappa\alpha}^\mu$ from muonic x-ray measurements are combined to provide

the model-independent mean-square charge radius, described in Ref. [93];

$$\langle r^2 \rangle = \frac{R_{\kappa\alpha}^\mu}{V_2^e} \quad (3.20)$$

In the fortunate cases where these measurements have been performed (such as for the element tin with 10 stable isotopes) the FS and MS factors can be extracted from measured values of $\delta\langle r^2 \rangle^{A,A'}$ and $\Lambda^{A,A'}$ using the linear relation,

$$\Lambda^{A,A'} = F_\lambda [\mu^{A,A'}]^{-1} \delta\langle r^2 \rangle^{A,A'} + k_{MS}. \quad (3.21)$$

Where the slope provide F_λ and the intersect directly gives the value of k_{MS} .

3.5.2 The field and mass-shift factors

The atomic factors F_λ and k_{MS} can be determined experimentally with the KP approach (Eqn. 3.21) by measuring IS of a selected atomic transition using precise measurements of mean-square charge radii. There are ten stable tin isotopes, presented in Tab. A.1, with accurately measured charge radii extracted from the combined analysis provided in Ref. [62]. A sample KP for IS measured in the $5p^2\ ^3P_0 \rightarrow 5p6s^3P_1$ atomic transition together with charge radii from Ref. [62] is presented as an example in Fig. 3.2(a). Normally the most accurately measured isotope is usually chosen as reference isotope A^{ref} . However, in this case where the charge radii of all isotopes are measured with equal precision the choice is not so clear. Different approaches for making a KP are discussed by Fricke *et al.* in [93] where a ladder technique is suggested where the IS between pairs of nearby isotopes are determined. Another approach is to choose a reference isotope based on the greatest KP range defined by $R = (y_{max} - y_{min})/y_{max}$ where $y_{max/min}$ is the largest and smallest y-values respectively.

A KP using different isotopes selected as reference should in principle give the same values for F_λ and k_{MS} . However, since the IS values are multiplied with atomic-mass dependant factors this is not the case. The extracted F_λ and k_{MS} from KP using different selected reference isotopes are presented in Fig. 3.2(b). This figure shows the scatter in the extracted F_λ and k_{MS} including the calculated KP range for the reference isotope used. The mean value for both F_λ and k_{MS} was chosen using the standard deviation of the scatter added in quadrature with the average sample error for the selected FS and MS factors. This is smaller albeit in agreement with the ladder method used in Ref. [62] for the determined F_λ and k_{MS} .

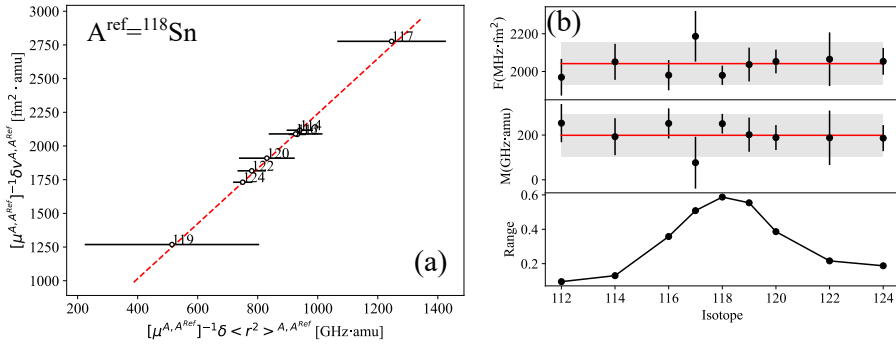


Figure 3.2: (a) shows a KP from measured IS of tin isotopes in the $5p^2\ ^3P_0 \rightarrow 5p6s^3P_1$ atomic transition, using IS and radii data provided in Ref. [62]. (b) the influence of the selected reference isotope on the extracted FS and MS factors, the average value presented with the red line was used in this work with error shown with grey shade.

3.5.3 Extracting nuclear moments and spin from the hyperfine structure

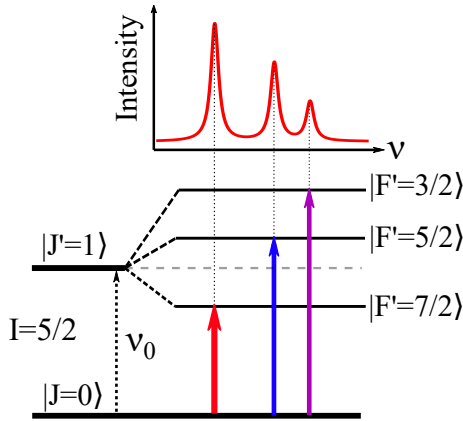


Figure 3.3: Transitions scheme to the hyperfine sub-states F' from $I = 5/2$ and $J' = 1$. The resulting hyperfine spectrum from the spectroscopy is shown in the inset graph indicating the difference in intensity of each resonance.

Electrons can resonantly excite between atomic hyperfine-structure levels when the atom is irradiated with light, see Fig. 3.3. The transition frequency between

the lower (l) J, F and upper (u) J', F' hyperfine structure levels result from the combined contribution from the magnetic dipole (Eqn. 2.11) and the electric quadrupole (Eqn. 3.16) interactions resulting in the expression;

$$\Delta\nu_{\text{hf}} = \nu_0 + \frac{A_{\text{hf}}^{(\text{u})} C'}{2} + B_{\text{hf}}^{(\text{u})} \frac{3C'(C' + 1) - 4IJ'(I + 1)(J' + 1)}{8IJ'(2I - 1)(2J' - 1)} \quad (3.22)$$

$$- \frac{A_{\text{hf}}^{(\text{l})} C}{2} - B_{\text{hf}}^{(\text{l})} \frac{3C(C + 1) - 4IJ(I + 1)(J + 1)}{8IJ(2I - 1)(2J - 1)}.$$

Where ν_0 is the frequency between fine-structure levels and C is defined earlier in Eq. 3.14. The allowed dipole transitions between hyperfine levels obey the transition rules,

$$\Delta F = 0, \pm 1 \text{ and } F = 0 \nrightarrow F' = 0. \quad (3.23)$$

The hyperfine structure spectrum resulting from spectroscopic measurements can be fitted using Eqn. 3.22 for values of the electric angular momentum and nuclear spin J, J', I . Once the hyperfine constants A_{hf} and B_{hf} are extracted from the fitted model the nuclear moments can be extracted by decoupling the atomic physics contribution accordingly:

$$g = \frac{A_{\text{hf}}}{A_0} \quad (3.24)$$

$$Q_s = \frac{B_{\text{hf}}}{q} \quad (3.25)$$

Where the hyperfine fields A_0 and q can be extracted either by measuring the hyperfine structure of a reference isotope with known nuclear moments or from calculations, as presented in the article added in Sec. 6.2.

Nuclear spin

When $I \geq J$ the fine-structure levels splits into $2J + 1$ hyperfine sub-states, if $I \leq J$ then the number of sub-states are directly determined by the nuclear spin $2I + 1$ down to $I = 0$ when the fine-structure is unperturbed. Therefore, the number of hyperfine structure peaks allows to directly determine the spin of the nucleus when $I < J$. For this reason fine-structure levels with higher total angular momentum number J are favorable for spin determination. However, the average transition intensity is reduced with the increased number of sub-states, transitions to the sub-states with lowest intensity can easily be lost in any present background signal. Furthermore, increased number of hyperfine levels increases the probability for peaks overlapping in the resulting spectrum. Extracting information from a convoluted spectrum with multiple peaks can prove particularly challenging.

Another way of distinguishing the spin from the hyperfine structure is to measure the relative intensities between hyperfine structure transitions. The intensity between hyperfine sub-levels relative to the underlying fine-structure intensity $S_{JJ'}$ is given by [111];

$$\frac{S_{FF'}}{S_{JJ'}} = (2F + 1)(2F' + 1) \left\{ \begin{matrix} F & F' & 1 \\ J' & J & I \end{matrix} \right\}^2 \quad (3.26)$$

These relative transition intensities can be compared to the experimentally observed for distinguishing the nuclear spin. However, it should be noted that relative intensities can often deviate in experimental measurements due to transitions being saturated or from other laser or beam related effects [112].

The electronic factors A_0 , q remain constant to a good approximation for the fine-structure levels of a selected transition, irrespective of the isotope, if hyperfine anomalies can be neglected. Then the ratio of the lower and upper hyperfine parameters should remain constant $\frac{A_{hf}^u}{A_{hf}^l} = \text{const}$ and $\frac{B_{hf}^u}{B_{hf}^l} = \text{const}$ for measurements along isotopic chains. This is particularly useful for determining the nuclear spin when $J \leq I$ since only the correct I for each isotope will reproduce a constant hyperfine ratio across the isotopic chain. The technique used for spin determination is discussed further in Sec. 7.4.

Chapter 4

Experimental technique for probing the hyperfine structure

Short-lived isotopes have been artificially produced for over a century from the bombardment of targets with high-energy particle beams from particle accelerators. These artificially produced nuclei could be separated chemically from the target material to study their properties. In the first decades, this limited measurements to elements with half-lives of a few minutes¹. The rapid development of 'online' radioactive ion beam (RIB) facilities based on electrostatic mass separation of isotopes, starting in the late 1960s allowed measurements of isotopes with tens of millisecond half-life, through rapid extraction and transportation as radioactive beams electrostatically accelerated to about 40 keV. One of these pioneering facilities was the ISOLDE (Isotope Separator Online DEvice) facility at CERN at the Swiss-French border near Geneva, where the work for this thesis was performed. Sensitive laser spectroscopy techniques have been developed hand in hand with these facilities, capable of performing measurements of hyperfine structures and isotope shifts of atomic systems containing exotic isotopes with interesting nuclear properties [24]. In this chapter the collinear resonance ionization spectroscopy (CRIS) technique is introduced together with the technical details necessary for performing the spectroscopic measurements on stable and radioactive tin isotopes in this work.

¹Often using the SRAFAP system -students running as fast as possible [113]

4.1 Spectroscopic techniques for resolving hyperfine structures of short-lived isotopes

Laser spectroscopic techniques share common standards for achieving accurate and reliable measurements of the hyperfine structures and IS:

- Resolution - In order to resolve isotope shifts and hyperfine structures of atomic states, a spectral resolution of <1 GHz is required with significantly higher resolution often needed in order to probe the minuscule perturbations of the atomic levels, which could be as small as <1 MHz, in particular for light elements.
- Signal detection efficiency and selectivity - The short-lived isotopes at the edges of nuclear existence which are often of interest can only be produced in minute quantities (few ions per second or less) together with unwanted contamination (isobars and molecular ions). It is therefore essential to achieve a high signal detection efficiency and isotope selectivity, along with excellent background suppression.
- Stability - To accurately determine the differences in IS with an accuracy of the order of a MHz, a stable environment for repeated measurements is required, providing both short-term stability and long-term stability of the experimental conditions (such as laser frequency, laser power, beam energy, intensity etc.) over the course of the measurements.

4.1.1 Collinear laser spectroscopy

The precision at which spectral lines can be resolved is often limited by the thermal velocity distribution of the atoms. The Doppler broadening effect of spectral lines (resulting from the velocity distribution) often constitutes the main limitation for resolving the hyperfine structures. When a gas in thermal equilibrium at temperature T is irradiated with light at the wavelength λ_0 as measured in the lab frame, the Doppler broadening of the observed spectral line can be calculated from the expression [114];

$$\delta\nu_D = \nu_0 \sqrt{\frac{8 \ln 2 k_b T}{m}} \approx \frac{214}{\lambda_0} \sqrt{\frac{T}{m}} \text{ GHz.} \quad (4.1)$$

As a result of the Maxwell-Boltzmann velocity distribution the Doppler broadening results in a Gaussian contribution to the spectral linewidth. Assuming a gas of tin atoms ($m=120$ amu) at 2500 K (the typical temperature

in the hot ISOLDE target environment, described in Sec. 4.2) probed with light at $\lambda_0 = 280$ nm, results in a Doppler broadening of the spectral lines of 3.5 GHz. Since the hyperfine structures are often < 5 GHz this makes the study of hyperfine interactions challenging. This broadening effect can be reduced by lowering the thermal motion of the ensemble using various cooling techniques [87]. Another approach is to utilize the velocity-bunching phenomena, by performing spectroscopy on fast moving beams, as first demonstrated in the early 1970s [115, 116]. When an ensemble of ions is accelerated to a velocity v (non-relativistic) the kinetic energy spread δE will be constant such that, $\delta E = mv\delta v = \text{const}$, thus leading to a reduced velocity spread δv if the beam is accelerated. Therefore, when an ensemble of ions are accelerated with the electrostatic potential U , reaching a velocity $v = \sqrt{2eU/m}$, the Doppler width from Eqn. 4.1 can be reformulated accordingly [114];

$$\delta\nu_D = \nu_0 \frac{\delta v}{c} = \nu_0 \frac{\delta E}{\sqrt{2eUm}c^2} \approx \frac{6921}{\lambda_0} \sqrt{\frac{\delta E}{mU}} \text{ GHz}. \quad (4.2)$$

Consequently, by probing a fast moving beam along the direction of motion (collinearly or anti-collinearly) results in a significantly reduced Doppler spread. Considering a bunch of tin ions being electrostatically accelerated to a kinetic energy of 20 keV for an energy spread of 5 eV, results in a Doppler linewidth of only 35 MHz. At this point, the Doppler broadening is often not the dominating contribution to the observed spectral linewidth.

As a direct consequence of Heisenberg's uncertainty principle the lifetime of the atomic level τ determines the uncertainty in the energy of the atomic level, resulting in a natural linewidth contribution;

$$\delta\nu_n = \frac{1}{2\pi\tau}. \quad (4.3)$$

For meta-stable states in tin with a typical short lifetime of 4 ns [107], the natural linewidth is calculated to be 40 MHz which is even greater than the expected Doppler broadening effect. Unlike the Doppler broadening which induces a Gaussian line profile, the natural linewidth contributes as a Lorentz distribution of the spectral profile, shown in Fig. 5.4.

The lifetime of the state is dependant on the interaction with laser field, which can influence the effective atomic state lifetime, resulting in a power broadening of the spectral linewidth [117];

$$\delta\nu_p = \delta\nu_n \sqrt{1 + \frac{I}{I_s}} \quad (4.4)$$

Where I_s is the intensity of the light where the transition is saturated, meaning the stimulated emission equals the transition probability between the atomic

levels. The laser power at which this occurs depends on the strength of the atomic transition given by the Einstein coefficient A_{ki} [118]. Therefore, in experiments the laser intensity is often reduced to just below this value to prevent power broadening of the spectral lines while maintaining high excitation efficiency [119].

It is important to note that in order to probe the fast moving atomic beam the frequency of the incoming light needs to be Doppler corrected to match the transition frequency in the rest frame of the atom,

$$\nu_{lab} = \nu_0 \sqrt{\frac{1 \pm \beta}{1 \mp \beta}}, \quad (4.5)$$

where $\beta = v/c$ with (+) for collinear and (-) for anti-collinear irradiation of the beam. This Doppler-shift provides an additional benefit when performing spectroscopy of elements containing many different isotopes: as the ion velocity is different for each isotope, the Doppler-shift separates the spectrum of different masses, thus reducing peak overlap and simplifying mass identification for heavier elements. The complete hyperfine structure of an atomic or ionic fine structure level can be probed either by tuning the frequency of the laser light directly (as done in this work) or by varying the beam velocity by changing the acceleration potential of the beam while maintaining a fixed laser frequency. When the frequency of the incoming light matches the transition frequency between two hyperfine levels, electrons will be excited from a lower state to an upper meta-stable state, which will subsequently decay with the release of a fluorescence photon. The fluorescent photons can be counted while scanning across the fine structure transition for revealing the hyperfine structure spectrum. Fluorescence detection has been developed many decades ago and is one of the detection methods successfully used at the COLLAPS beamline at ISOLDE [120].

4.1.2 Resonance ionization spectroscopy

An electron can be step-wise excited to higher atomic levels when irradiated with subsequent laser steps before the decay of the excited atomic state(s). This step-wise excitation process can bring the electron above the ionization potential (IP), thereby ionizing the atom. There are three common approaches for laser resonance ionization, as schematically demonstrated for a simplified two-step resonance ionization scheme in Fig. 4.1. In Fig. 4.1(a) the electron that is resonantly excited to a high lying meta-stable state can be non-resonantly brought above the ionization potential if the incoming photon has sufficient

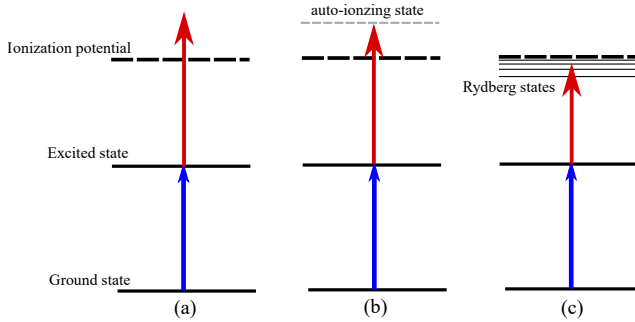


Figure 4.1: (a) display two step scheme with a resonant step together with non-resonant laser ionization, scenario (b) show subsequent resonant ionization by exciting into an auto-ionizing state near the IP and in scenario (c) the second laser step brings the electron into a high lying Rydberg state where it can be collisionally ionized or brought above the IP via field ionization.

energy. The cross-section for non-resonant ionization into the continuum is low, this process therefore requires a light source with high power density. Atomic states lying above the lowest ionization threshold with short lifetimes can spontaneously decay by electron emission. Excitation to these so called auto-ionizing states (Fig. 4.1(b)) have a significantly increased cross-section (several orders of magnitude) as compared to non-resonant ionization. The electron can also be resonantly excited into a high-lying Rydberg states, which appears just below the IP as seen in Fig. 4.1(c). Electrons in these states can be detached when the atomic potential is perturbed by an external electric field (known as field-ionization process), or by collisions or black-body radiation. The resonance ionization process allows for selective ionization of an element from their characteristic energy spectrum. It can thus be understood that the element selectivity can be enhanced by increasing the number of resonant steps in the laser ionization scheme and by reducing the laser linewidth of each step [24]. This element specific selectivity with resonance ionization is particularly useful for purifying beams containing many different elements produced at isotope separator facilities, and is regularly used at the Resonance Ionization Laser Ion Source (RILIS) at ISOLDE for selective ionization of elements of interest [121].

Spectroscopic measurements can be performed in the ion source by scanning the wavelength of one of the resonant excitation steps: when the frequency matches a transition the electron is step-wise excited until detached from the atom. The ion can be easily separated from the other atoms in the gas with electrostatic fields, directing the ions into an ion detector where they can be counted. This resonance ionization spectroscopy (RIS) procedure can

achieve over 50% efficiency and is often performed close to the source where the exotic isotopes are produced for reducing transport efficiency losses. The method is then known as in-source spectroscopy [122]. The dominant sources of background come from non-resonantly ionized events, such as surface ionization. This resonance ionization spectroscopy technique suffers from the Doppler broadening of the ions in the hot target, as discussed earlier, which makes spectroscopy of hyperfine structures and isotope shifts limited to heavy elements or elements with large hyperfine splittings, as only low resolution measurements can be achieved with a typical resolution of the order of a few GHz [111].

4.1.3 Collinear Resonance Ionization Spectroscopy (CRIS)

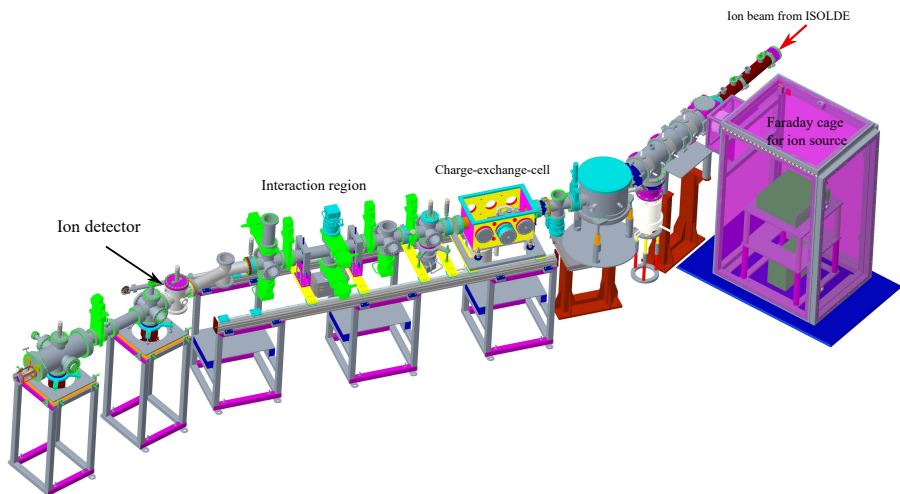


Figure 4.2: Overview of the CRIS beamline at the ISOLDE facility. Courtesy of Andrew Smith.

Resonance ionization spectroscopy can be performed in a collinear geometry for achieving high-resolution, while maintaining a high ion detection efficiency necessary for probing the most exotic atoms; this procedure was first demonstrated in 1991 [123] and has more recently been perfected at the collinear resonance ionization spectroscopy (CRIS) beamline at the ISOLDE facility [124, 125, 126], shown in Fig. 4.2. The state-of-the-art CRIS technique is capable of performing laser spectroscopic measurements with a resolution down to 20(1) MHz of exotic isotopes produced at yields of only tens of ions per second, reaching total efficiencies up to 1% from production to detection²[99, 126, 127].

²The CRIS efficiency is described in Sec. 4.9.

This makes the technique particularly suitable for performing high-resolution laser spectroscopic measurements of the most exotic tin isotopes, of interest in this thesis. A description of the CRIS technique follows, guided by the schematic overview of the CRIS beamline in Fig. 4.3:

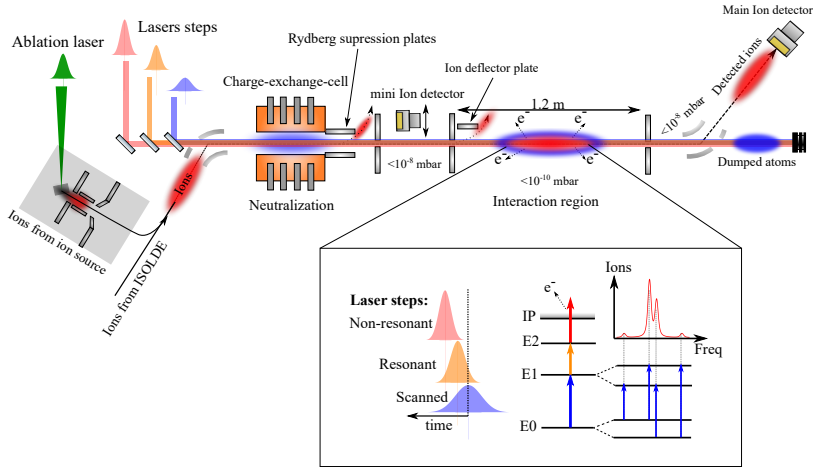


Figure 4.3: Schematic overview of the CRIS beamline. Ions produced from either ISOLDE or the laser ablation ion source are sent into a charge-exchange-cell which neutralizes the beam before entering the interaction region. Pulsed lasers overlap with the atomic bunch in the interaction region. Here, lasers resonantly excite and ionize the atoms when the lasers are on resonance. By counting the ions while scanning across the hyperfine structure using a narrowband laser reveal the hyperfine structure, as shown in the inset figure. Another ion detector after the charge exchange cell can be inserted for measuring the neutralization fraction or for time-of-flight (TOF) measurements. More details are described in the text.

Mass selected and bunched ion beams containing the radioactive or stable isotopes of interest can be injected into the CRIS beamline on-line from ISOLDE at a set beam energy up to 60 keV (described in Sec. 4.2). Additionally, an ion source attached to the CRIS beamline can provide pulsed or continuous ion beams containing stable elements (described in Sec. 4.7). The injected ions are steered and focused using electrostatic electrodes after a 34° bend into a heated charge-exchange-cell (CEC) containing alkali metal vapour. Ions passing the CEC are partially neutralized into different atomic states from the charge-exchange process, described in greater detail in Sec. 4.4. The partially

neutralized beam passes charged plates generating an electrostatic field of 5000 V/cm perpendicular to the beam direction, thereby removing remaining ions from the neutralized beam while field-ionizing any high-lying long-lived Rydberg states that might also be populated in the charge exchange process [126]. The beam continues through an iris into a differential pumping region, held at an average pressure of 10^{-8} mbar. In this region a MagneToF ion detector is attached to a retractable actuator. This detector is used for characterizing beam properties, such as neutralization efficiency (discussed in Sec. 4.8) and for performing time-of-flight (TOF) measurements, see appendix A.2. The beam then passes a deflector plate generating a perpendicular field of 1000 V/cm for removing residual collisionally ionized ions, before entering the interaction region. The interaction region is a 1.2 m section of the beamline maintained at an ultra-high vacuum of 10^{-10} mbar using a set of two turbomolecular ion pumps. This reduces the chances for collisional ionization of the atomic beam. Here, a set of laser pulses are collinearly overlapped and synchronized in time and space with the atomic bunch at 100 Hz duty cycle (details on the synchronization are found in Sec. 4.7). A narrow-bandwidth (<60 MHz) pulsed laser is scanned across a selected hyperfine structure transition from a low-lying atomic state populated in the charge-exchange process using a resonance ionization scheme tailored for the element of interest. The versatile laser systems used in the experiments are described in Sec. 4.5. A three-step ionization scheme is demonstrated in the inset in Fig. 4.3. If the lifetime of the atomic states involved in the transition is sufficiently long, then the resonant laser steps can be delayed with respect to each other to prevent line distortion effects of the probed transition [112, 128]. The resonantly ionized ions in the bunch are electrostatically steered with electrostatic plates past a 20° bend into the main MagneToF ion detector which counts ions on an event-per-event basis. By bending the resonantly ionized beam away from the remaining non-ionized beam particles, very low background count rates can be achieved in the ion detector. By counting the number of ion events at different measured laser frequencies a near Doppler-free measurement of the hyperfine structures is performed. While this is the normal mode of operation at CRIS the versatile design allows for modular attachments after the 20° bend, by retracting the main ion detector. The resonantly ionized beam can be e.g. steered into a decay spectroscopy setup for performing α -decay tagging on isomeric states [129] or β -decay assisted laser spectroscopy for reducing the influence of isobaric contamination in the RIS beam [130], with many future opportunities on the horizon.

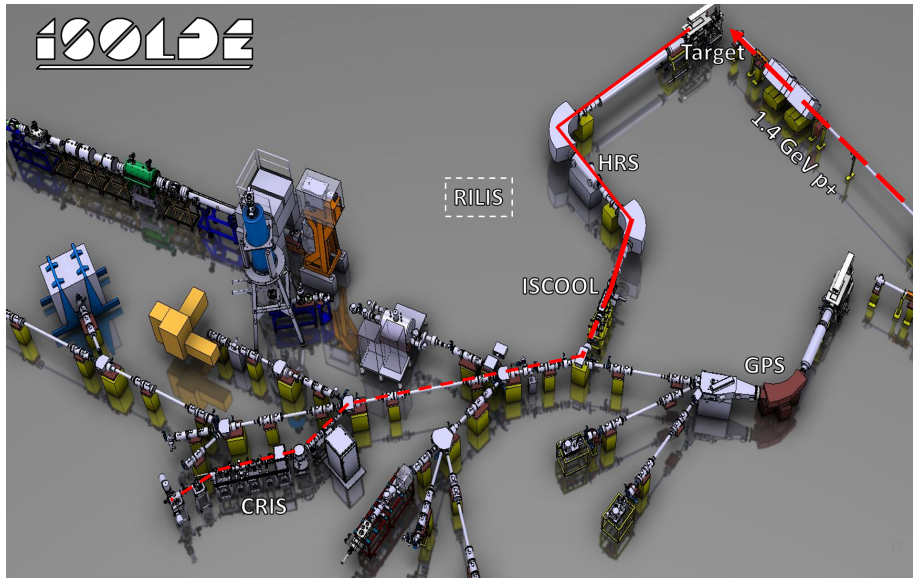


Figure 4.4: Overview of the low energy section of the ISOLDE facility. 1.4 GeV protons accelerated from the PS booster are directed onto a heated target. The produced short-lived isotopes generated within the target are extracted and sent through HRS mass separator magnets before being injected into the ISCOOL cooler buncher. The bunched radioactive beam is finally transported to the CRIS beamline.

4.2 Radioactive isotope production at ISOLDE

The ISOLDE radioactive beam facility provides versatile production and mass separation of short-lived radioactive isotopes, which are sent to a large set of experimental stations located within the ISOLDE experimental hall [131]. The facility is divided into a low energy area for experiments accepting ion beams at energies up to 60 keV and a high-energy sections after the REX/HIE-ISOLDE post-accelerator for experiments requiring ion beam energies up to 10 MeV/u [132]. The CRIS beamline is located in the low energy section of the ISOLDE hall, shown in Fig. 4.4. Bunched beams of radioactive ions of a selected isotope are separated at ISOLDE and electrostatically steered into the CRIS beamline where spectroscopic measurements can be performed.

In order to produce radioactive isotopes at ISOLDE protons were first accelerated

using LINAC2³ up to 50 MeV before being injected into the proton synchrotron booster (PSB), which further accelerates the beam up to an energy of 1.4 GeV [133]. The PSB generates a proton beam with discrete bunches containing up to 3×10^{13} protons per pulse of 900 ns pulse length with 1.2 s duty cycle. Approximately half of these bunches are directed to the ISOLDE facility. At ISOLDE the protons bombard a 4 cm in diameter and 20 cm long cylinder filled with a target material where short-lived isotopes can be generated from spallation, fragmentation or fission reactions within the tailor-made target [134]. In order to generate neutron-deficient tin isotopes a porous LaC_x target material was chosen, as the fragmentation of Lanthanum ($Z = 57$) facilitates the generation of neutron-deficient species in the $A = 100$ region of the nuclear landscape [135]. The target material is heated to temperatures as high as 2000°C for enhancing the diffusion and effusion of the produced radioisotopes out of the target. The produced isotopes escape the heated target material into a separately heated transfer line leading the radioactive gas into an ion source region. There, the elements of interest can be ionized using a variety of techniques such as arc discharge, surface ionization or resonance ionisation using RILIS [136]. Selective resonance ionization using RILIS was used to resonantly ionize the generated tin atoms using an efficient three-step ionization scheme from the $5s^25p^23P_0$ atomic ground state to the $5p9s^2P_2$ auto-ionizing state [137], which provided an estimated ionization efficiency of roughly 20% [138]. The generated ions are electrostatically accelerated up to 40 keV and steered into the High Resolution Separator (HRS) magnet. This mass separator is composed of two tunable C magnets seen in Fig. 4.4, a 90° and a 60° magnet together with collimator slits which provide a mass resolution of $\frac{\Delta m}{m} = 6000$ [131]. The mass selectivity using HRS together with the element selectivity of RILIS, should produce a rather pure isotopic beam. However, isobars with a low IP might also be ionized through surface ionization, as well as molecular ions, leading to contamination which can for some masses constitute a significant fraction of the beam. For very exotic isotopes, this contamination is often significantly greater than the yield of the isotope of interest. For beams of tin isotopes a high isobaric contamination of surface ionized indium is expected due to the significantly lower IP as shown in Fig. 4.5.

³This linear accelerator was decommissioned shortly after the experiment was performed in 2018.

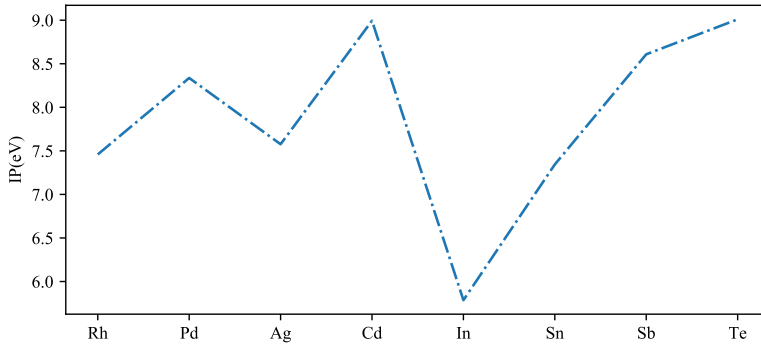


Figure 4.5: IP of elements near tin. The low IP of indium means it is likely to contribute to surface ionized isobaric contamination in tin beams produced at ISOLDE.

The mass filtered ion beam from HRS is then injected and decelerated into an 800 mm long gas-filled segmented linear Paul trap (called ISCOOL) [139], shown schematically in Fig. 4.6. The radioactive ions are electrostatically trapped within a helium gas environment which allows the ion beam to lose energy from thermal collisions with the helium buffer gas, held at a pressure of about 0.1 mbar (further details are found in Ref. [131]). The release of the bunches from ISCOOL is synchronized with the CRIS duty cycle of 100 Hz, meaning the ions can be trapped for up to 10 ms before release. This cooler buncher is essential for performing efficient CRIS measurements as it reduces duty cycle losses from the pulsed nature of the CRIS technique. After release the ion bunch is guided using electrostatic bender plates and focusing elements into the CRIS detection setup where spectroscopic measurements can be performed. Based on measured yields down to ^{105}Sn using a LaC_x target, the production yields of neutron-deficient tin isotopes were extrapolated [140], as shown in Tab. 4.1. The observed yields in this work are presented later in Chp. 7.

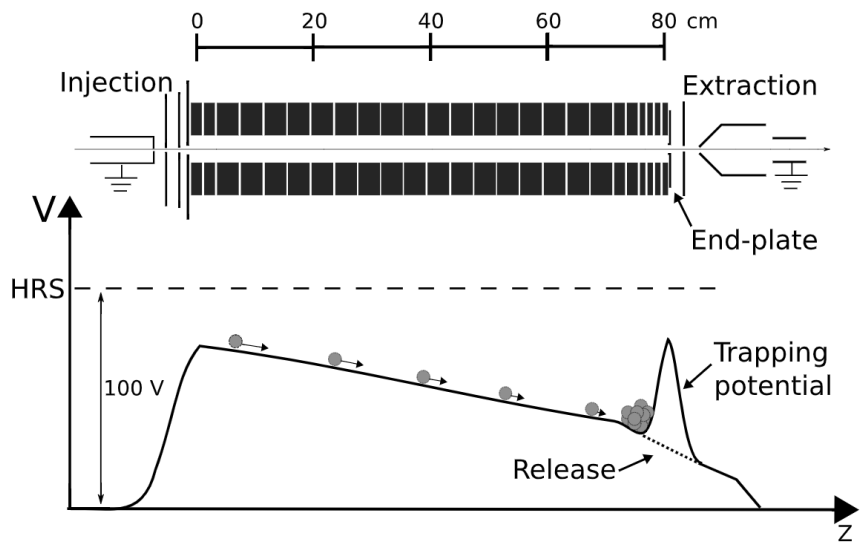


Figure 4.6: Cross-section of the ISCOOL buncher for generating bunched radioactive beams at ISOLDE. Taken from Ref. [139].

Table 4.1: Estimated production yields of the radioactive tin isotopes relative to charge of proton current (typically 1.4 μ A) bombarding a LaC_x target at ISOLDE [140]. Extrapolated values are in indicated in bold font.

Element	A	Half-life	Yield at ISOLDE(ions/ μ C)
Sn	101	1.7 s	<1.0E+01
Sn	102	4 s	<1.0E+02
Sn	103	7 s	<1.0E+03
Sn	104	21 s	1.0E+04
Sn	105	31 s	1.0E+05
Sn	106	115 s	1.6E+06
Sn	107	2.90 m	6.0E+06
Sn	108	10.30 m	1.4E+08
Sn	109	18.0 m	1.0E+09
Sn	110	4.11 h	1.8E+09
Sn	113m	21.4 m	6.0E+09

4.3 The laser ablation ion source

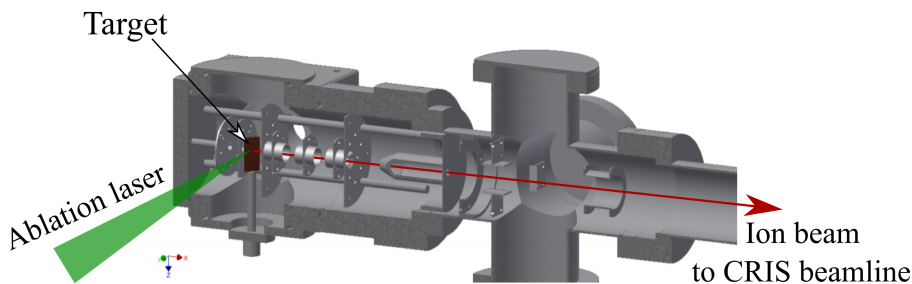


Figure 4.7: Cross-section of the laser ablation ion source used at CRIS. Pulsed laser light is focused onto a metallic tin target angled at 45° , laser ablation generates ions which are focused and electrostatically accelerated into the CRIS beamline.

The CRIS ion source (seen in Fig. 4.7), facilitates versatile production of ion beams of stable elements off-line (when no protons are available) using a variety of ion source techniques, such as surface ionization or laser ablation [141]. The surface ionization source is composed of a 4 cm long and 2 mm tantalum (melting temperature of 3290 K) capillary tube attached in one end to a small container filled with salts, containing the element of interest such as KCl for K^+ production [142]. The metallic container and capillary are resistively heated to temperature high enough to dissociate the molecule and generate K^+ from surface ionization. This generates a continuous stream of ions which can be electrostatically accelerated and steered into the CRIS beamline. This ion source is however not suitable for the generation of tin ions since the IP of tin (7.34 eV) is significantly greater than the work function of tantalum at 4.5 eV.

Another mode of operation is by performing laser ablation of solid targets [141]. This technique provides an extraordinarily versatile way of producing a wide range of stable elements including molecular ions using either compound or metallic targets. In this mode of operation, 532 nm pulsed laser light produced from a Litron LPY Nd:YAG system (described in Sec. 4.5) is focused onto the target angled at 45° . The laser is focused to an approximate spot size of 1 mm in diameter generating a fluence of roughly $> 0.5 \text{ J/cm}^2$ at the focal point, see Fig. 4.7. The laser interaction with the surface is a particularly complex process which is highly dependant on the power densities involved [143]. Much simplified, the laser pulse energy results in rapid heating of the target surface. The strong laser field rips the electrons from the atoms, thereby generating an expanding plasma plume of atoms and ions. The light electrons are rapidly

accelerated outwards from the interaction point leaving the heavier positively charged ions of different charge states. The Coulomb repulsion between the positively charged ions results in a prompt burst of ions with a large energy spread of 0.1-1 keV [144]. This prompt component is in the order of the laser interaction time, which is around 10 ns. This release is followed by a slow thermal release of atoms and ions from the hot surface, lasting up to a few microseconds, which is significantly cooler than the prompt component and typically follows a Maxwell-Boltzmann velocity distribution [145]. The generated ion bunch containing ions of varying charge states experience the electrostatic potential within the chamber, floating at a variable voltage of 0 to +30 kV. The electrostatic field accelerate the ions into an Einzel lens together with a set of steering electrodes which focuses the beam into the CRIS beamline after a 90° electrostatic bend; details of the beam optics are found in [126, 141]. The 1+ ions are filtered by the electrostatic beam transport through the beamline. It is however possible for different charge states to slip through as contamination.

An extensive investigation was performed using an indium target by studying the release of ions at different applied extraction fields applied at the target position, described in Ref. [109, 141]. This work indicated a reduced velocity spread of the generated bunch below <3.5 eV with a field gradient less than 0.01 V/m applied at the ablation target. The low extraction voltage increases the TOF of the ion bunch but reduces energy spread, enabling high-precision spectroscopic measurements of ablated species using the CRIS technique. Many different metallic targets have been successfully used for ion beam production using the ion source such as In, Ga, Zn, Cu and Sn. The ion production yields using laser ablation of metallic targets is initially high but stabilizes with time, fluctuations of about 7% were observed on a 10 s timescale after 30 minutes of use [109]. Experiments using this source are generally performed at least 1h after starting the ablation laser, lasting up to 8h without changing the target position. Ideally, rotating targets would be used for increasing the target lifetime and enhancing yield stability. A metallic tin target for the laser ablation ion source provides an almost ideal source of tin ions for off-line experiments. This is because natural tin contains the most amount of stable isotopes of all elements. There are seven even spin-zero isotopes $^{112,114,116,118,120,122,124}\text{Sn}$ and three spin-half isotopes $^{115,117,119}\text{Sn}$ at the disposal for spectroscopic measurements, presented together with their natural abundance in Tab. A.1 in the appendix. This source was used for probing the stable tin isotopes in this work, discussed further in Chp. 6.

4.4 Ion beam neutralization

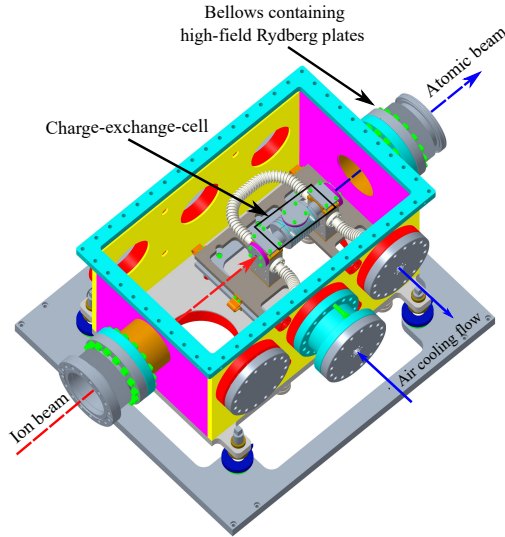


Figure 4.8: CAD overview of the section of the CRIS beamline containing the CEC. Courtesy of Andrew Smith.

Performing CRIS on atoms is considerably easier than on ions. This is partly because the energy required to remove a second electron from an ion is much greater than the energy required to ionize the atom, thus requiring many more resonant laser steps and/or deep UV lasers. Furthermore, it is significantly easier to separate neutral atoms from 1^+ ions than it is to separate the 2^+ from the 1^+ charge state.

A charge-exchange-cell (CEC) for neutralizing the ion beam is therefore added to the CRIS beamline. The CEC, seen in Fig. 4.8, is held within a vacuum chamber at a pressure below 10^{-7} mbar. The center of the CEC is filled with approximately 20 g of alkali metal, usually metallic sodium or potassium. A temperature of 350°C can be reached within the cell using resistive heater elements, which melt the alkali metals ($T(\text{Na})=98^\circ\text{C}$ and $T(\text{K})=64^\circ\text{C}$) generating alkali vapour within the cell. The ends of the CEC are cooled with a constant airflow through connected bellows in order to maintain a cycle of evaporation and condensation which maintain the alkali vapour within the cell. The vapour pressure can be maintained at 10^{-4} mbar for $>200\text{h}$ without significant evaporation losses from the cell.

When a beam of tin ions in the ionic ground state enter the CEC the loosely bound electrons in the alkali metal $A(0)$ can be captured by the passing tin ions, resulting in neutralization into an atomic state k accordingly,

$$\text{Sn}(0)^+ + A(0) \rightarrow \text{Sn}(k) + A(0)^+ + \delta E. \quad (4.6)$$

Where $\delta E = I_{\text{Sn}} - I_A$ is the so called energy defect given by the energy difference between the ionization potential of the two elements [146]. The first ionization potential of tin at $I_{\text{Sn}}=7.344$ eV is significantly greater than that of both sodium $I_{\text{Na}}=5.139$ eV or potassium $I_{\text{K}}=4.341$ eV taken from [147]. This results in a large energy defect, allowing high-lying meta-stable states k in Sn to be populated in the charge-exchange-process. While the total neutralization cross-section for the incident tin beam is difficult to calculate accurately, the relative cross-section for neutralization into the atomic states k is possible to simulate with reasonable accuracy [148, 149]. A simulation code by A. Vernon et al. [148] based on the semi-classical impact-parameter approach by Rapp and Francis [146] was used together with spectroscopic data from [147] to simulate the relative population in different atomic states after the charge-exchange process. The relative atomic population simulated assuming a 40 keV tin ion beam interacting with either sodium or potassium vapour is presented in Fig. 4.9 and 4.10. The neutralized beam then travels approximately 120 cm from the CEC into an interaction region before spectroscopic measurements are performed. A TOF of 4.73 μs allows short-lived atomic states to decay into lower lying states which redistributes the relative population; this is shown in black as the final relative population of tin. When using sodium neutralizer a considerable population is expected in the high-lying meta-stable state $5p^2\ ^1S_0$ at 17425 cm^{-1} which is close to the energy defect at $\delta E = 17783\text{ cm}^{-1}$. The greater energy defect when using potassium neutralizer $\delta E = 24219\text{ cm}^{-1}$ favors a higher occupancy in the short-lived $5s6p$ states with nanosecond life-times [107]. These states quickly decay down to the $5s^2$ states before the beam reaches the interaction region. A small fraction of the neutralized atoms are populated in high-lying Rydberg states. The proximity to the IP make atoms populated in these dark states particularly susceptible to collisional ionization which contributes to the background signal. By applying an external electrostatic field F [V/cm] atomic states occupied at the energy E_n can be field-ionized [150];

$$E_n - \text{IP} = -5.97\sqrt{F}\text{ cm}^{-1}. \quad (4.7)$$

An electric field of 5000 V/cm was applied perpendicular to the beam path after the CEC which field-ionized atoms populated in states 70 cm^{-1} below the IP, while also removing any residual ions from the beam before entering the interaction region.

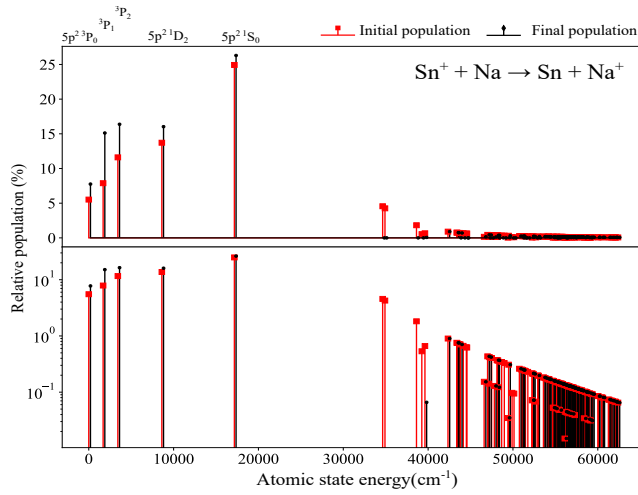


Figure 4.9: The simulated atomic population after the neutralization of a 40 KeV Sn ion beam passing the CEC containing Na vapour. The initial atomic population within the CEC as well as the final population after the beam reaches the interaction region are presented. The logarithmic scale reveal the population in higher lying states.

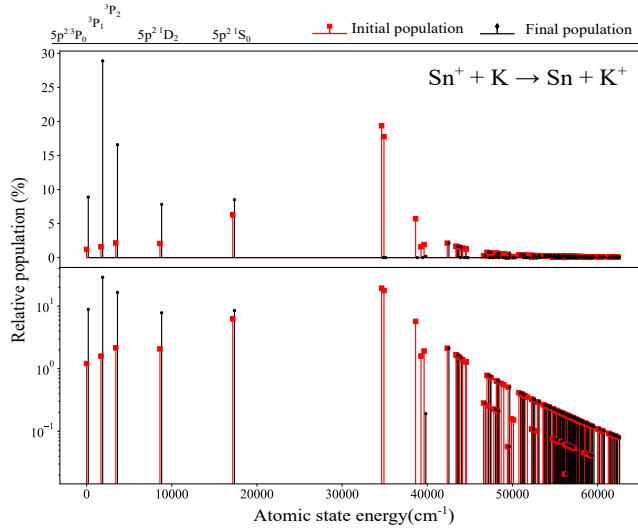


Figure 4.10: Simulated atomic population in Sn after charge exchange with K vapour under similar conditions as Fig. 4.9.

4.5 Laser systems

An impressive arsenal of both pulsed and continuous-wave (CW) laser systems of broad and narrow linewidth are available at the CRIS experiment for performing versatile high-precision spectroscopic measurements of the elements produced at ISOLDE. The combination of tunable pulsed dye lasers (PDL) and titanium-sapphire (Ti:Sa) laser systems and their higher harmonics give access to transitions ranging from 1000 nm down to 207 nm, necessary for exploring different resonance ionization schemes for different elements. The laser setup used at CRIS for performing spectroscopy of tin in this work is displayed in Fig. 4.11.

Accurate scanning across the hyperfine structure transitions requires precise tunable lasers. Two CW Ti:Sa lasers systems are available for this purpose: The Matisse Ti:Sa laser by Sirah and SolsTiS Ti:Sa laser by M-Squared. These CW lasers are mode-locked to a single mode by fine tuning the effective length of the optical resonator cavity using frequency-selective elements such as a birefringent filter (Bifi) and etalons, using stepper motors or piezo actuators [151]. Once locked, the laser output can be tuned with sub-MHz steps with a power output up to 10 W across the Ti:Sa laser range of roughly 770-920 nm, a more detailed description of these laser systems can be found in [151, 152].

While CW lasers offer excellent spectral resolution they are not suitable for probing the hyperfine structure transitions at CRIS due to the optical pumping into dark states which reduces the efficiency of a measurement [153, 154]. Pulses of laser light can be produced from the CW output by chopping, using fast switching of a Pockels cell with polarization-selective optics [155]. However, this technique often does not provide the photon density required to saturate weaker transitions. Furthermore, higher-harmonic generation of CW lasers requires expensive and complex additional modules that only produce adequate powers for the 2nd harmonic.

One solution is to use the narrowband CW laser to seed a pulsed injection-seeded Ti:Sa laser system for generating pulsed laser light with high laser fluence while maintaining a narrow bandwidth (Fourier-limited to 20 MHz) at the seeded laser frequency; a more detailed description of this laser system can be found in [156, 109]. The injection-seeded Ti:Sa laser system used at CRIS is pumped with 532 nm pulsed laser light at a repetition rate of 1 kHz generated from a LEE Laser LDP-100MQ frequency doubled Nd:YAG laser. The LEE Nd:YAG laser system provides an internal trigger for the Q-switch at 1 kHz repetition frequency, this trigger is used as the master trigger for the CRIS experiment. This system is used to pump multiple pulsed Ti:Sa systems by dividing its output using half-wave plates and polarizing beam-splitter cubes.

A sum-harmonics unit for tripling the laser frequency, described in [157], was used to gain access to transitions in tin available in the third harmonic of Ti:Sa laser light, at 250-290 nm. Initially, the second harmonic is generated after passing a BiBO (bismuth triborate BiB_3O_6), both the fundamental and second Harmonic are split into separate paths. This is done to tune the polarization of the fundamental light using a $\lambda/2$ to phase match with blue for Type I mixing in BBO (β -barium borate $\beta\text{-BaB}_2\text{O}_4$) crystal for generating the third harmonic, the crystal angles are specified for the desired frequency. Typically, 200 mW fundamental Ti:Sa laser light typically generates 2 mW output power in the third harmonic. The UV laser beam is shaped using a set of cylindrical lenses and focused to an approximate beam diameter of 5 mm into the CRIS beamline at a distance of about 25 m using a set of mirrors. Residual second harmonic light is diminished to less than 1% of the third harmonic into the beamline, with about 50% transport efficiency of the third harmonic into the CRIS beamline.

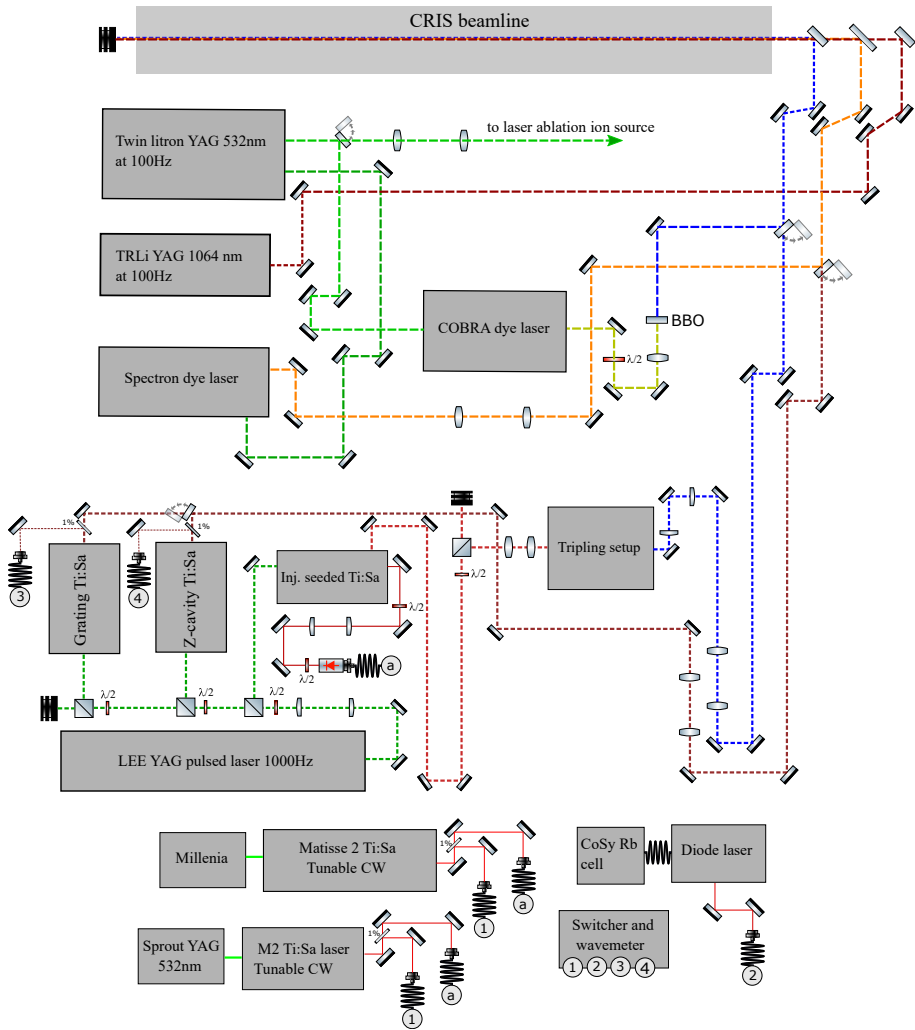


Figure 4.11: Overview of the laser setup at CRIS (see text for details).

Two broad bandwidth Ti:Sa laser systems are pumped at 1 kHz using the LEE laser, a Z-cavity, presented in [158], and a Grating Ti:Sa, presented in [159]. The Z-cavity Ti:Sa laser provides broadband light with a spectral linewidth of 6 GHz in the range 680-960 nm depending on the used mirror set. The wavelength is manually set by rotating a Bif and tuning the angle of an etalon. It is rarely used for scanning long ranges (searching for transitions) because birefringent filters cause distinct modes that are not continuously tunable across.

On the other hand, the grating Ti:Sa laser is capable of rapid and continuously tunable frequency scanning within the full Ti:Sa range from 700-940 nm using a motor-controlled grating [159]. This broadband laser has a pulse energy up to 200 μ J per pulse and a laser linewidth of 2 GHz.

A laser table next to the CRIS beamline is dedicated to a set of dye laser systems and Nd:YAG lasers operating at 100 Hz. These pulsed lasers define the duty cycle of CRIS measurements. A dual-head Litron pulsed Nd:YAG laser provides 532 nm light at 50 mJ per pulse with 8 ns pulse duration from two independent laser heads. This laser provides pump light for two dye laser systems, Cobra dye laser from Sirah and a Spectron Spectrolase 4000 pulsed dye laser (PDL). When ablation studies are performed one of the laser outputs from the dual-head Litron is focused onto the ablation target in the laser ablation ion source described in 4.7. A flip mirror is used to redirect the light for pumping the PDL or the COBRA dye laser when taking beam from ISOLDE.

A Litron Nano TRLi HR 250-100 pulsed Nd:YAG laser system provides 4 ns pulses of up to 250 mJ per pulse at 1064 nm for achieving high-ionization efficiency, and is usually used as the last non-resonant step in resonance ionization schemes studied in tin. Its higher harmonics, 532 nm and 355 nm, can be generated by attaching additional modules to the output head. The COBRA and PDL dye laser systems allow access to wavelengths not accessible using Ti:Sa lasers and their harmonics. Different dyes dissolved in ethanol are used as lasing medium in these lasers. In this work Rhodamine 6G and DCM dyes were used for generating light in the ranges between 560 to 610 nm and 610 to 665 nm respectively. The PDL laser provides broadband light of approximately 10 GHz and was used for intermediate resonance steps. The broad bandwidth is needed to cover any hyperfine structure of the transition to ensure high resonance ionization efficiency. A neutral density filter was used for reducing the output of the laser light into the beamline to reduce power broadening effects discussed earlier. The COBRA dye laser system provides reduced linewidth down to roughly 2 GHz. This light could be doubled using a BBO crystal for generating 280 nm light for performing medium-resolution scanning of atomic transitions. A CRIS measurement of the $281 \text{ nm } ^1S_0 \rightarrow ^1P_1$ transition was performed by scanning the frequency doubled light from the COBRA laser with Rhodamine 6G dye medium, or with frequency tripled light from the injection-seeded laser, shown in Fig. 4.12. This spectrum indicates the difference between a high-resolution measurement using the injection-seeded laser achieving a linewidth of 200 MHz compared with 3 GHz using doubled light from the COBRA dye laser.

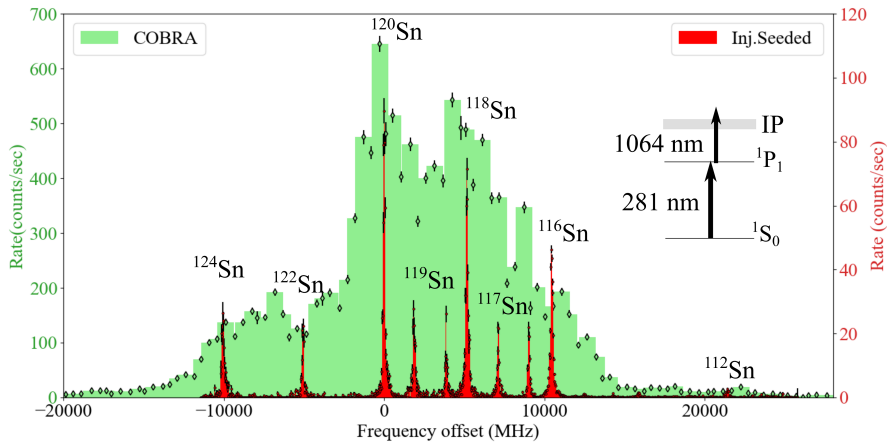


Figure 4.12: CRIS measurement of stable tin isotopes (Frequency offset: $35544.3156 \text{ cm}^{-1}$). The $^1S_0 \rightarrow ^1P_1$ transition indicated in the inset figure is scanned using the 2nd harmonic from the COBRA dye laser and with tripled light from the injection-seeded Ti:Sa laser system. Roughly 3 GHz resolution is achieved using the COBRA while a resolution of 200 MHz is achieved when scanning with the tripled injection-seeded laser light.

4.6 Frequency measurements

Accurate and precise monitoring of the laser frequency when scanning a hyperfine structure transition is essential for performing a CRIS measurement. Two Fizeau-based interferometers by HighFinesse were used for precise frequency measurements [160]. A WSU-2 wavemeter was used for measuring the laser frequency of the narrow bandwidth lasers used for scanning the hyperfine structures. This wavemeter has a specified accuracy of 2 MHz when calibrated within 2 nm of the measured transition frequency, allowing measurements of frequencies in the range 330 to 2250 nm. Since the scanned transition often is outside this spectral range, for example when using sum harmonics generation with the Injection-seeded laser system, the fundamental light frequency is measured instead of the third-harmonic output. The measured frequency is then multiplied by 3 when processing the data. The laser light is captured after pickoff mirrors near the outputs of the laser systems and directed using an optical fiber into a four-channel switch which allows simultaneous measurement of up to four lasers. The measured frequency is subject to drifts caused by change in environmental conditions in the laser lab, such as temperature, humidity and pressure [161]. In order to correct for any drift in the wavemeter response the

light from a stabilized diode laser (DLC DL PRO 780 by Toptica Photonics AG) is measured simultaneously. This laser diode is connected with a compact saturated absorption spectroscopy setup (CoSy FC-Cs+Rb+K) used for locking the laser to Doppler-free hyperfine-absorption lines. The hyperfine transition $5S_{1/2}(F=2) - 5P_{3/2}(F'=3)$ of ^{87}Rb , with an absolute frequency measured to 384 228 115 208(17) kHz [162], was used to lock the laser diode in the experiments in this thesis. The long term drifts of the locked diode is expected to be below <1 MHz.

Additionally, a WS6-600 wavemeter was used to monitor the frequencies of the pulsed dye lasers. This wavemeter has an absolute accuracy of 600 MHz within the range 375-800 nm which is sufficient for monitoring the frequency of the broadband laser light.

4.7 Timing synchronization

The pulsed nature of the CRIS experiment requires precise synchronization in time in order to carefully overlap the laser pulses with the probed atomic bunch. The time synchronization of all systems at CRIS are performed using a set of Quantum Composers 9520 digital-delay pulse generators for accurate time control with 250 ps timing resolution and <50 ps jitter. A pulse diagram indicating the timings used during a CRIS experiment is shown in Fig. 4.13. The 1 kHz logic internal trigger pulse for the LEE Laser Q-switching is used as the master trigger for the experiment. Since the LEE laser system provides pump light for the injection-seeded laser system (used for narrowband scanning of the hyperfine structure) its pulse timing is fixed. The Nd:YAG lasers next to the CRIS beamline operate at 100 Hz which set the duty cycle limit for the experiment. The pulse delay generators were therefore set to trigger these lasers on one in every ten pulses from the master trigger. During experiments utilizing ISOLDE, the delay generator provides a trigger pulse initiating the ISCOOL release. The delay is set according to the distance from ISCOOL to the interaction region in the CRIS beamline (approximately 22 m) and the velocity of the ion bunch, corresponding to 86.7 μs for ^{120}Sn at 40 keV beam energy. When the laser ablation ion source is used it is typically set around 45 μs before for tin at 20 keV acceleration voltage, and adjusted for optimal overlap with the laser pulses in the interaction region. The timings of all lasers were adjusted for optimizing the efficiency of the resonance ionization process.

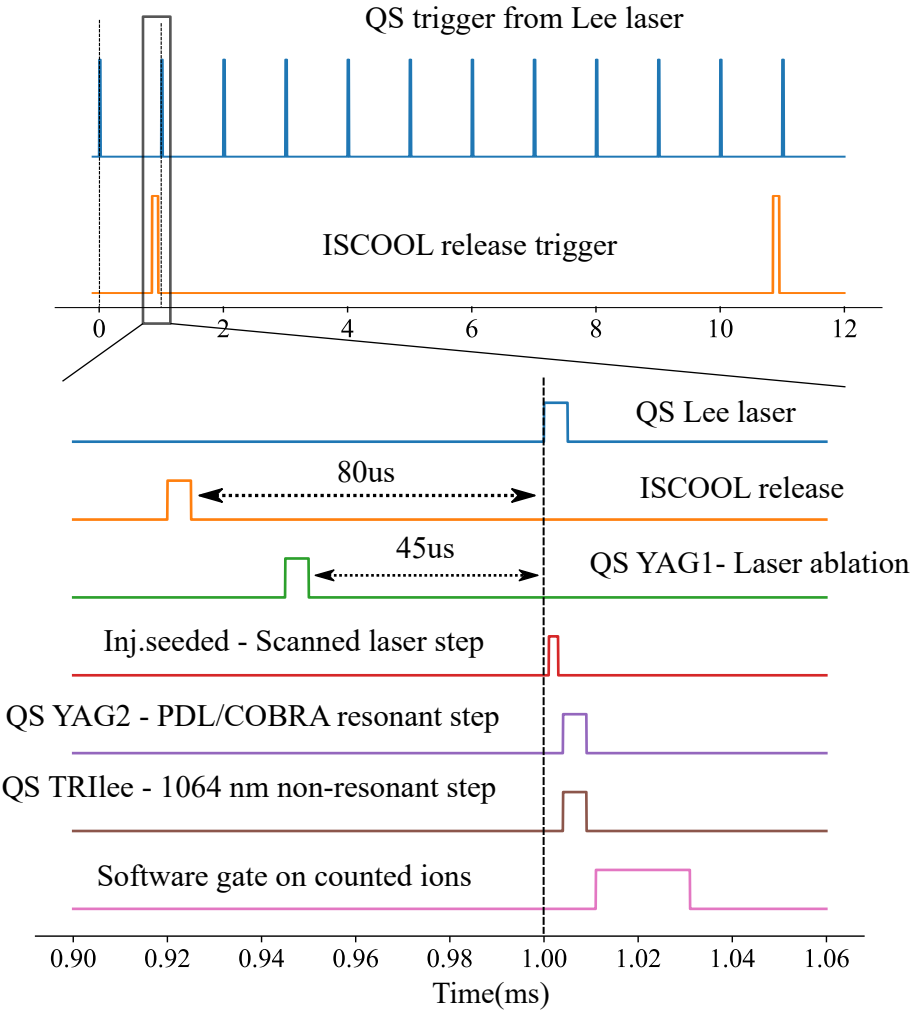


Figure 4.13: Timing diagram of the synchronization during a CRIS measurement.

4.8 Ion detection

The resonantly ionized ions are detected using a DM291 ETP MagneTOF ion detector which has sub-ns FWHM timing resolution for event by event ion counting. Arriving ions pass a thin metallic grid and are impinged on a stainless steel plate covered in a dynode material coating. Electrons generated from

the impact are directed with electric and magnetic fields towards additional dynodes which amplifies the generated charge. This charge is converted using a charge sensitive preamplifier to a measurable output signal of roughly -30 mV. The bias voltage is set between +2500 to +4000 V with increased bias voltage with increased age of the detector. These detectors provide a fast voltage recovery after a burst with a linear ion counting response up to $6 \cdot 10^{10}$ cps [163]. The signal is further amplified in a fast amplifier before being sent into a Time-to-Digital Converter (TDC, model Cronologic TimeTagger-42G) data-acquisition card where an internal trigger threshold is set. Every triggered event is registered with a time value (t) from a set start time (t_0) with 500 ps time resolution. t_0 is synchronized with the laser pulses at 100 Hz generated by the pulse delay generator. The TOF for every ion event is registered by setting t_0 to the ISCOOL release time or the Q-switch of the ablation laser. This information can be used to discriminate against non-resonantly ionized events such as noise generated by the Q-switch and in certain cases facilitate further processing of the data, which can enhance the quality of the acquired spectra as discussed in the Sec. 5.5.

4.9 Detection efficiency of a CRIS measurement

The total detection efficiency ϵ of ions released from ISCOOL to detection at CRIS is given dominantly by the following contributions,

$$\epsilon = \epsilon_t \epsilon_n \epsilon_p \epsilon_l \epsilon_d \quad (4.8)$$

where ϵ_t is the beam transport efficiency from ISCOOL to the CRIS beamline. This is difficult to measure accurately due to the different gain of the Faraday cups used to measure the beam current but is expected to reach up to 70%. ϵ_n is the fraction of the beam that is neutralized following the CE interaction after the CEC. This is measured using a mini MagneToF ion detector that can be inserted in the region after the CEC (see Fig. 4.3). The detected beam current can be measured under two conditions: (i) the neutralized beam intensity is measured by deflecting the remaining ions using a biased deflector plate just after the CEC, (ii) the full beam intensity (ions + atoms) is measured by removing the bias on this plate. The ratio of the full beam and the neutralized beam is used to determine the neutralization efficiency. This procedure was used to estimate a neutralization efficiency for tin beams of 20(10)% when using either sodium or potassium vapour within the CEC. ϵ_p corresponds to the relative population within the probed atomic state after neutralization and

subsequent decay into the final state population configuration, as presented in subsection 4.4. Therefore, an atomic state with significant final population should be chosen to maximize the CRIS efficiency. Simulations presented in Sec. 4.4 indicate the populations in each atomic state which varies from roughly 10% up to 30% for the five lowest lying atomic states in tin, see Fig. 4.9 and 4.10. The laser-atom efficiency ϵ_l can in theory reach near 100% assuming the transitions are saturated and the laser-beam overlap is perfect. Finally, the detection efficiency ϵ_d is assumed around 80% using the MagneToF ion detector, where only minor losses are expected from scattering on a grid in front of the detector. With all these contributions considered a total efficiency of roughly 1% is expected in an optimal scenario during the experiment. While the maximized efficiency is highly element dependant due to the contributions from ϵ_p and ϵ_n an efficiency of 1% has been achieved in previous experiments at CRIS [128, 164].

4.10 Data acquisition systems

All data at CRIS are acquired using a Python based data acquisition (DAQ) system CRISTAL (CRIS laser Tuning, Acquisition and Logging) which enables direct communication between different computers connecting the measuring devices such as wavemeters and ion detectors. This program also enables the controlling of the scan speed of the laser scanning by setting the scan speed and scanning range. Data are continuously recorded during a measurement until the scan range has been reached. The data are saved in comma-separated values (CSV) format datafiles with a common timestamp for every data row which allow the merging of the data. More information on the data acquisition can be found in, Ref. [153].

Chapter 5

Data handling and analysis

This chapter describes the treatment of the raw data obtained from a CRIS measurement. The necessary corrections and filtering of the data required to refine the quality of the spectrum are also described. Furthermore, the acquired TOF information facilitates further Doppler correction of spectroscopic measurements of beams produced from the laser ablation ion source are described here.

5.1 Raw data treatment

A complete CRIS measurement is contained within four CSV format datafiles. The measured wavenumbers using the two wavemeters are saved in separate files for each wavemeter. The measured ion events with corresponding TOF for each ion event relative to start tagger, and the ISCOOL platform voltage measured continuously, are saved in two separate files. Each measured value has a global timestamp which allows merging the different data sets into a single datafile. The merged datafile contains the following information for every row in the dataset: t (timestamp), n (ion events), dt (ion TOF), $wn1$ (wavenumber of the scanned laser step), $wn2$ (wavenumber of the intermediate resonant laser step), wn_{ref} (wavenumber of the locked reference laser diode) and V (ISCOOL platform voltage). This raw data can be easily manipulated using Python together with NumPy [165] and Pandas [166] processing packages. The tagger DAQ card registers each ion event with a TOF value in units of 500 ps, dt is therefore divided by 2 to convert the values into nanoseconds. When no ion event is registered within 10 ms of the start trigger, a row is created with $dt=-1$,

these rows are not removed since they are required to accurately calculate the ion count rate. The timestamp t is used to calculate the time difference between each row in the data set and added as a separate column Δt . The summed ion events and Δt are used for calculating the count rate.

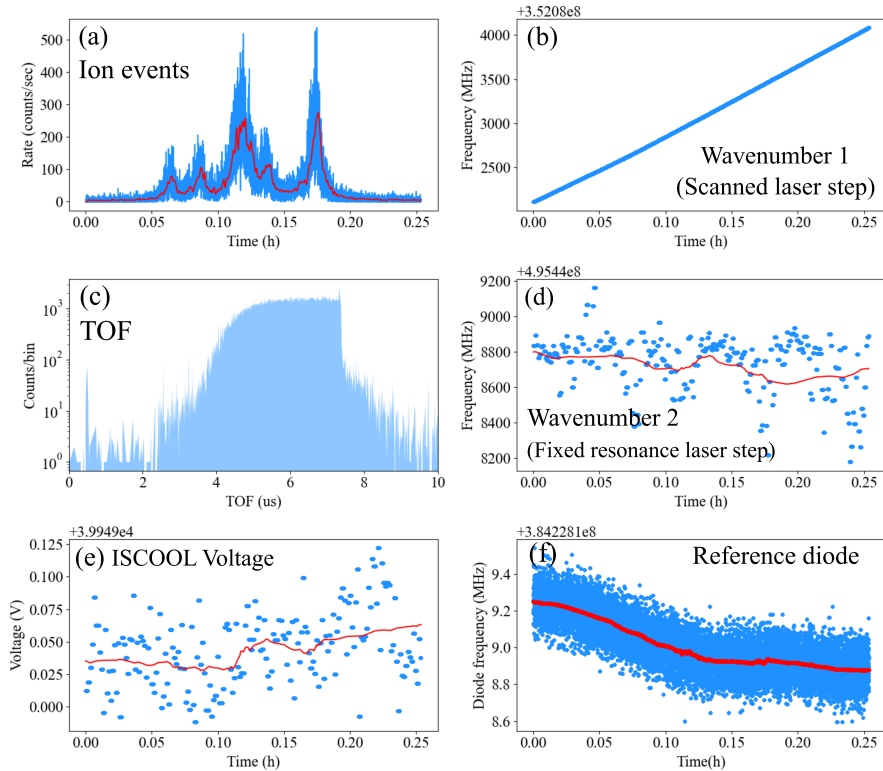


Figure 5.1: Overview of the acquired raw data from a single CRIS measurement of ^{107}Sn produced at ISOLDE. The figures (a,c,e,f) are shown together with their 5s moving average displayed in red. Details of these figures are explained in the text.

The raw data acquired from a 17 minutes CRIS measurement scanning the hyperfine structure of ^{107}Sn , is shown in Fig. 5.1. The number of ion events registered within a 1 s timeframe (ion count rate) is recorded over the duration of the measurement, seen in Fig. 5.1(a). This reveals the ions collected over time while the frequency of the first laser step is scanned across the hyperfine structure, Fig. 5.1(b). A TOF spectrum of the captured ion events can be

created by binning with respect to dt , with typical bin-size of 100 to 500 ns depending on the statistics, Fig. 5.1(c)¹. The data can be gated in TOF to reduce non-ion related background events outside the ion bunch arrival time. This is particularly useful for removing electric noise generated from the Q-switching lasers nearby or ionization from scattered UV light.

The measured frequency of the PDL, set to a fixed intermediate resonance step is also monitored, as seen in Fig. 5.1(d). A 30 s moving average is used to clarify any drift occurring during the measurement. The broad linewidth of over 10 GHz means the small drift of 200 MHz is of little concern over the course of a measurement, as this is within the accuracy of the WS6-600 wavemeter used to measure this frequency (see Sec. 4.6). The wavelength of the intermediate resonance step was adjusted in between measurements to account for any long-term drifts. The ISCOOL platform voltage and the reference diode frequency were monitored during a measurement, as shown together with their 5s moving average in Fig. 5.1(e) and (f).

In rare cases artifacts were observed in the count rate, see Fig. 5.2. These artifacts show a sudden drop in count rate with zero counts registered for up to a few seconds, followed by a sudden burst. The integrated number of counts within the burst corresponds to the missing counts within the drop. The events within the burst all have equal TOF which suggests the artifact is caused by a computer related issue. In most cases the artifacts were small enough to be filtered out during the binning procedure, but in some cases this could not be done. In these cases the faulty data was removed manually from the spectrum.

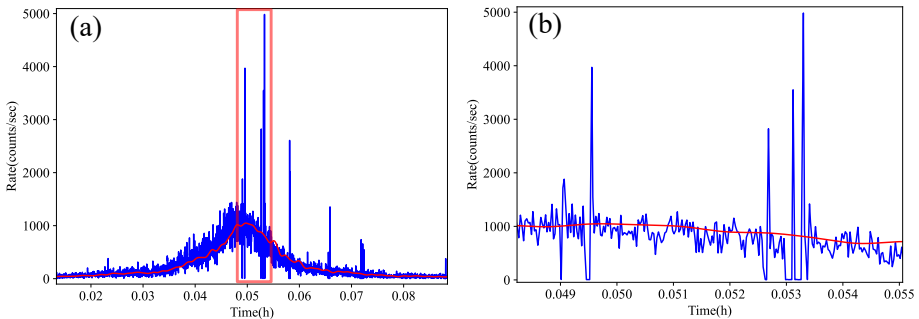


Figure 5.2: A sample spectrum revealing sudden bursts in the data seen in Fig. (a) together with a highlighted region with a zoom in Fig. (b). These were identified as a computer related issue and the faulty data was removed from the spectrum.

¹The shape of this distribution is described in greater detail in Sec. 7.4.

5.2 Data corrections and binning procedure

The measured wavenumber using the wavemeter is subject to drift over the course of the measurement due to changes in environmental conditions such as humidity and temperature. This is why simultaneous measurements are performed using a reference diode laser locked to a well known reference hyperfine structure transition, with the known 'true' wavenumber ν_{True}^{Ref} (details are found in section 4.6). The ratio of the experimentally measured reference value $\nu_{measured}^{Ref}$ and the 'true' value is proportional to any other measured wavenumber using the same wavemeter accordingly;

$$\frac{\nu_{Measured}^{Ref}}{\nu_{True}^{Ref}} = \alpha \frac{\nu_{Measured}}{\nu_{Corr}}, \quad (5.1)$$

where α accounts for a possible non-linearity in the wavemeter response. Thus a measured wavenumber can be corrected using the expression,

$$\nu_{Corr} = \frac{\nu_{Measured}}{1 + \alpha \left(\frac{\nu_{Measured}^{Ref}}{\nu_{True}^{Ref}} - 1 \right)} \quad (5.2)$$

The factor α can be determined by performing repeated CRIS measurements of a selected atomic transition together with continuous measurements of the reference wavenumber. The value of α can be found by minimizing the scatter of multiple measurements $\sigma(\nu_{True})$ by varying α ; this procedure is used in Sec. 7.2. These drifts are often not significant over short time periods. A 5 hour long CRIS measurement is shown as an example in Fig. 5.3 which indicates less than 1 MHz drift over the course of the measurement. A 30 s moving average of the measured reference wavenumber (shown in red) is presented in the figure.

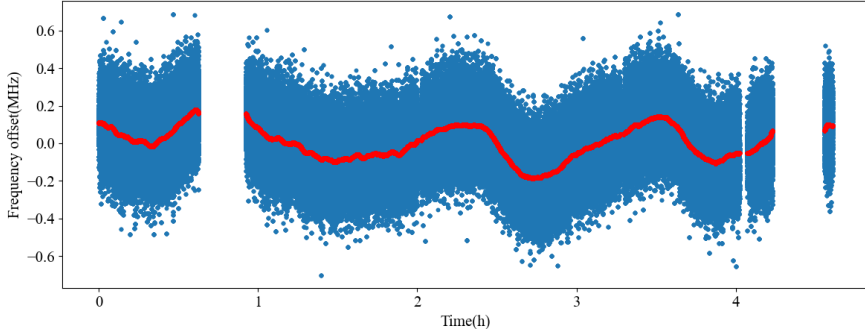


Figure 5.3: Merged measurements of the reference diode frequency offset over approximately 5h. The red line indicate the 30 s moving average.

The drift corrected wavenumber ν_{Corr} needs to be converted into the n^{th} -harmonic of light sent into the beamline, $\nu_{Lab} = n \cdot \nu_{Corr}$. Furthermore, Doppler correction is required to obtain the wavenumber of the light in the reference frame of the probed atom ν_{Atom} . This correction can be performed using Eqn. 4.7 knowing the acceleration voltage U (ISCOOL platform or the ablation ion source voltage) and the atomic mass m^2 of the elements in the collinerly overlapped atomic beam,

$$\nu_{Atom} = \nu_{Lab} \sqrt{\frac{1+\beta}{1-\beta}} \quad \text{where} \quad \beta = \sqrt{1 - \frac{m^2 c^4}{eU + mc^2}}. \quad (5.3)$$

Once the measured wavenumbers have been converted to the rest frame of the atom with appropriate drift corrections, the data can be binned with respect to wavenumber to generate a spectrum. The spectra in this work are normally presented with respect to frequency which is obtained from the wavenumber from $f_{Atom}[\text{MHz}] = \frac{10^4}{c} \nu_{Atom}[\text{cm}^{-1}]$.

The selected bin size varies depending on the conditions and statistics acquired during the measurement, as well as on the observed linewidth. It is typically selected to between 10 to 20 MHz. The influence of the selected bin size on the fitted spectrum is discussed in Sec. 7.7. The scanned frequency region of the CRIS measurement is subdivided with the selected bin size. Since the scan speed is often below 5 MHz/s there is a minimum of 200 data rows within each bin (10 ms per row if no event is registered). The total time spent within each bin is given by the summed contribution of all events Δt_i . The ion count rate

²The masses used for the selected tin isotope are given in Tab. 7.1.

can therefore be calculated from the summed contribution of all registered ion events $\sum_i n_i$ divided by the total time $\sum_i \Delta t_i$, with uncertainty given assuming poisson statistics:

$$y_i = \frac{\sum_i n_i}{\sum_i \Delta t_i}, \quad \sigma(y_i) = \frac{\sqrt{\sum_i n_i}}{\sum_i \Delta t_i}. \quad (5.4)$$

Similarly, the mean frequency is selected within each bin, with the error given by the standard deviation of the registered frequency values contained within the bin.

5.3 Fitting procedure

The SATLAS (Statistical Analysis Toolbox for Laser Spectroscopy) Python package was used for fitting the hyperfine structure spectrum, details of this package are found in [167]. This program generates a hyperfine structure model based on initial conditions such as centroid position, FWHM and the hyperfine constants A_{hf} , B_{hf} with fixed conditions for the line profile and spins I, J, J' . A summed model can be created when more than one isotope or isomer is present in the spectrum, in these cases the FWHM is shared for all hyperfine structure models.

Typically, a model is fitted to the acquired data using the least χ^2 method where the following function is minimized,

$$\chi^2 = \sum_i \frac{(y_i - f(x_i))^2}{e_i^2} \quad (5.5)$$

Here, y_i is the measured value with uncertainty e_i and $f(x_i)$ is the model predicted value. The goodness-of-fit for a selected model compared with data is commonly measured in terms of $\chi_r^2 = \chi^2/NDOF$, where NDOF is the number of degrees of freedom. When $\chi_r^2 = 1$ this means that the deviation of the model is as expected given the data variance. This value is greater than 1 if the model does not fit the data or if the data uncertainties are underestimated. Multiple measurements are often performed, if these measurements are independent then the weighted-mean value is used with standard weights given in terms of the statistical uncertainty σ_i accordingly, $w_i = \frac{1}{\sigma_i^2}$. The weighted-mean and its uncertainty are calculated from,

$$\bar{x} = \frac{\sum_{i=1}^n w_i x_i}{\sum_{i=1}^n w_i}, \quad \text{with, } \sigma_{\bar{x}} = \sqrt{\frac{1}{\sum_{i=1}^n w_i}}. \quad (5.6)$$

Where χ^2 is given by

$$\chi^2 = \frac{1}{(N-1)} \sum_{i=1}^n w_i (x_i - \bar{x})^2; \quad (5.7)$$

In order to account for systematic uncertainties not accounted for in the data the uncertainty is scaled accordingly $\sigma_{\bar{x}}^{(scaled)} = \sigma_{\bar{x}} \sqrt{\chi_r^2}$ when $\chi_r^2 > 1$.

5.4 Line profile

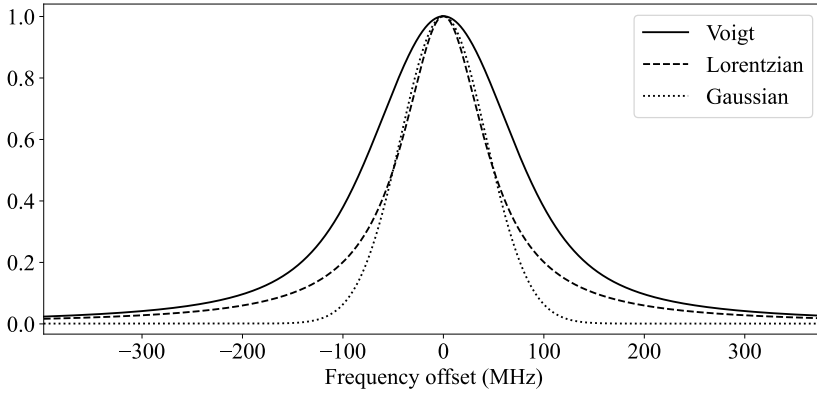


Figure 5.4: Different peak profiles used for fitting the hyperfine spectra. The Lorentzian(L) and Gaussian(G) profiles are simulated with 100 MHz linewidth and the Voigt profile with ($\sigma_L = 100$ MHz and $\sigma_G = 100$ MHz).

There are several different spectral line profiles considered in this work, see figure 5.4. However, the most common profiles are the Gaussian and Lorentz and the convolution of both, called the Voigt profile. The Gaussian contribution to the line profile stems from the statistical distribution of velocities of the atomic bunch, resulting in Doppler broadening effects. The Gaussian function is described by,

$$g(x; A, \mu, \sigma) = \frac{A}{\sigma\sqrt{2\pi}} \exp\left(-\frac{1}{2} \frac{(x - \mu)^2}{\sigma^2}\right). \quad (5.8)$$

Where A is the peak intensity, μ is the centroid position and σ is the FWHM of the peak. Moreover, there is the natural line-width resulting from the finite

lifetime of the atomic state and the power broadening contribution, described by the Lorentz function;

$$f(x; A, \mu, \gamma) = \frac{A}{\left[1 + \left(\frac{x-\mu}{\gamma}\right)^2\right]} = A \left[\frac{\gamma^2}{(x-\mu)^2 + \gamma^2} \right]. \quad (5.9)$$

Where 2γ is the FWHM. Both these contributions are combined in spectroscopy resulting in a convolution between the two functions named the Voigt profile,

$$V(x; \sigma, \gamma) = \frac{\text{Re}[w(z)]}{\sigma\sqrt{2\pi}}, \text{ where } z = \frac{x + i\gamma}{\sigma\sqrt{2}}. \quad (5.10)$$

The FWHM σ_v can be well approximated with;

$$\sigma_v \approx 0.5346\sigma_L + \sqrt{0.2166\sigma_L^2 + \sigma_G^2}. \quad (5.11)$$

There are various effects which can alter the symmetry of the spectral line such as Stark-shift effects and charge-exchange interactions, discussed in more detail in Ref. [112]. The peak profile in this work showed no sign of asymmetry, investigated in Sec. 7.2.

5.5 TOF correction procedure

The captured TOF data enables further corrections of the acquired hyperfine spectrum [107, 109, 141]. The frequency and TOF data acquired from a CRIS measurement of stable tin isotopes (Tab. A.1) produced from the laser ablation ion source at 20 keV beam energy, is presented in Fig. 5.5. This 3D representation shows the frequency spectra as it evolves over the TOF, the projection of the ion counts with respect to frequency and TOF are presented on each axis.

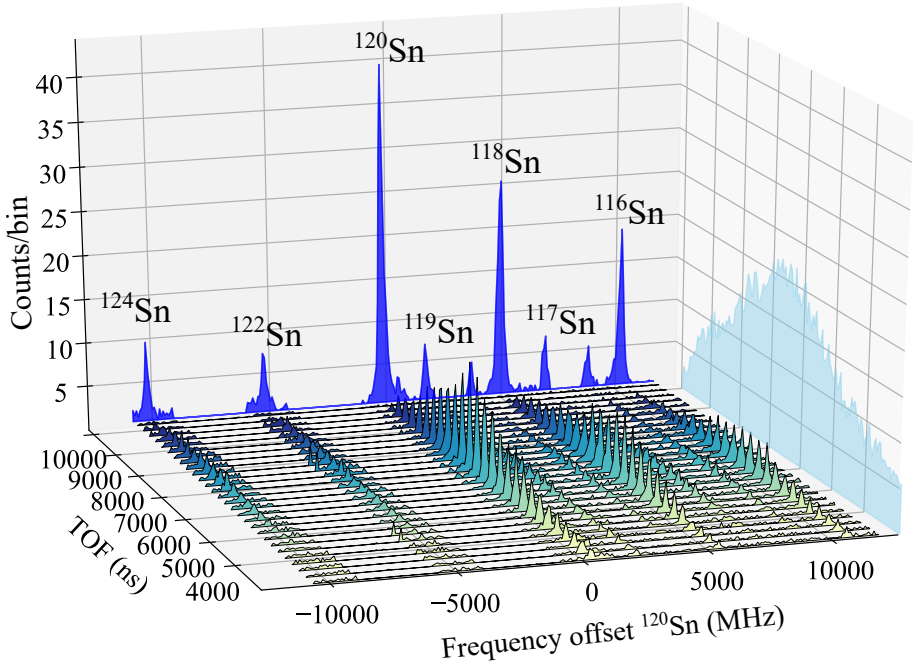


Figure 5.5: This 3D plot presents the TOF, frequency and ion events acquired from a CRIS measurement of stable tin isotopes when scanning the $5p^2\ ^1S_0 \xrightarrow{281\text{nm}} 5p7s\ ^1P_1$ transition and ionizing using 1064 nm light. The total ion counts are projected on the frequency and TOF axis. This information allows further correction for improving the quality of the spectra (see details in text).

The TOF distribution of each tin isotope can be studied by gating on the events in the identified resonance peak. An asymmetric TOF distribution with a tail extending towards shorter TOF is observed when gating on the events for each separated mass, see Fig. 5.6(a). A quasi Maxwell-Boltzmann distribution was used to fit this TOF distribution, formed from the convolution of a normal distribution function (Eqn. 5.4) and a cumulative distribution function (CDF) accordingly;

$$\text{CDF: } \Phi(x) = \frac{1}{2} \left[1 + \text{erf} \left(\frac{x}{\sqrt{2}} \right) \right] \text{ where } \text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt \quad (5.12)$$

$$\rightarrow F(x) = A\Phi(x) \cdot g(x) + C \quad (5.13)$$

Where A and C are constants describing the amplitude and background offset. This distribution function represented the data well as seen in the fitted TOF spectra for the even-tin isotopes masses, Fig. 5.6(a). A slightly longer tail extending towards shorter TOF was observed for the heavier tin isotopes. The reason for this is likely due to the greater fraction of non-resonant background events at $^{124,122}\text{Sn}$ due to the lower relative abundance of these isotopes, see Fig. 5.5. This tail is likely caused by the scattered ions from the prompt ion release from the ablation process, see appendix A.2. Due to the asymmetry of the distribution the median of the distribution was used for determining the TOF of each isotope. The median for each mass is indicated with a dashed line in Fig. 5.6(a). The TOF with respect to mass is presented in Fig. (b). While the TOF is expected to be proportional to the square root of the mass (as presented in appendix A.2) a near linear trend in TOF is observed within this mass range, with a slope of 190(30) ns/amu. The events from the different masses can be adjusted such that the median of the distribution lies along the same TOF, which allow the narrowing of the time-gate for reducing unrelated ion events [107].

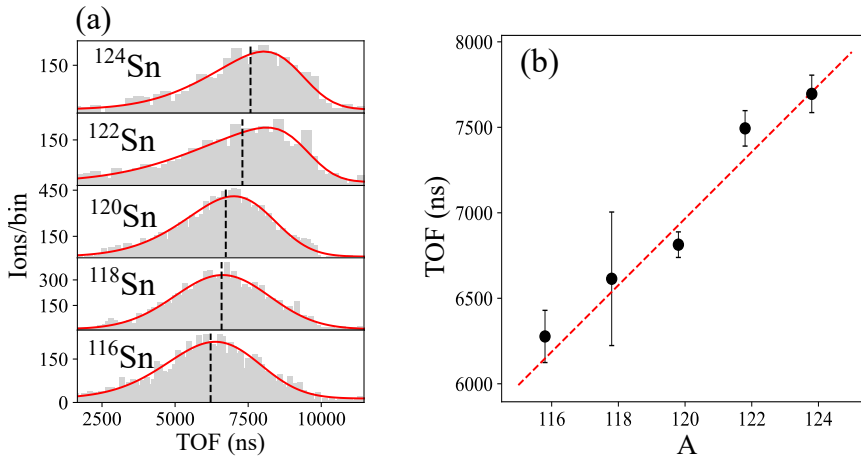


Figure 5.6: The TOF distribution of the different masses are fitted in (a) the median of the TOF distribution versus mass is fitted with a linear function in (b). The TOF increases to a good approximation linearly within the small mass range.

Despite the limited TOF resolution, with a FWHM of 2.5 μs , the perspective

provided by the additional information of TOF allows further identification of compound targets. This could prove useful when spectroscopic measurements are performed on ablated species containing many different compounds, which are resonantly ionized in the same spectrum, by determining the relative mass difference from the TOF of each resonance peak.

The velocity distribution of the probed atomic bunch gives rise to a shift in frequency with respect to the TOF, caused by the Doppler effect. This shift is observed when performing spectroscopic measurements of tin beams produced from the laser ablation ion source. This is revealed from the change in resonance frequency when gating on different TOF slices, see the example for ^{120}Sn in Fig. 5.7(a). The centroid frequency with respect to TOF was determined by fitting the resonance of each TOF slice, the 'banana'-shaped centroid vs TOF is revealed, see Fig. 5.7(b). The shape of this trend can be understood from the Doppler effect (Eqn. 5.2) with $\beta = \frac{v}{c} = \frac{\Delta L}{c\Delta t}$, which result in the expression,

$$\nu(t) = \nu_0 \sqrt{1 + \frac{2\Delta L}{c\Delta t - \Delta L}} = \nu_0 \left(1 + \frac{\Delta L}{c\Delta t} + \dots\right) \approx \nu_0 + \frac{k}{\Delta t} \quad (5.14)$$

Hence, to first order the trend $\nu(t)$ can be approximated with the simple expression $\nu_0 + \frac{k}{\Delta t}$. This expression is seen fitted to the centroid frequencies with respect to TOF for ^{120}Sn , indicating excellent agreement as presented in Fig. 5.7(b) together with the residuals in Fig. 5.7(c).

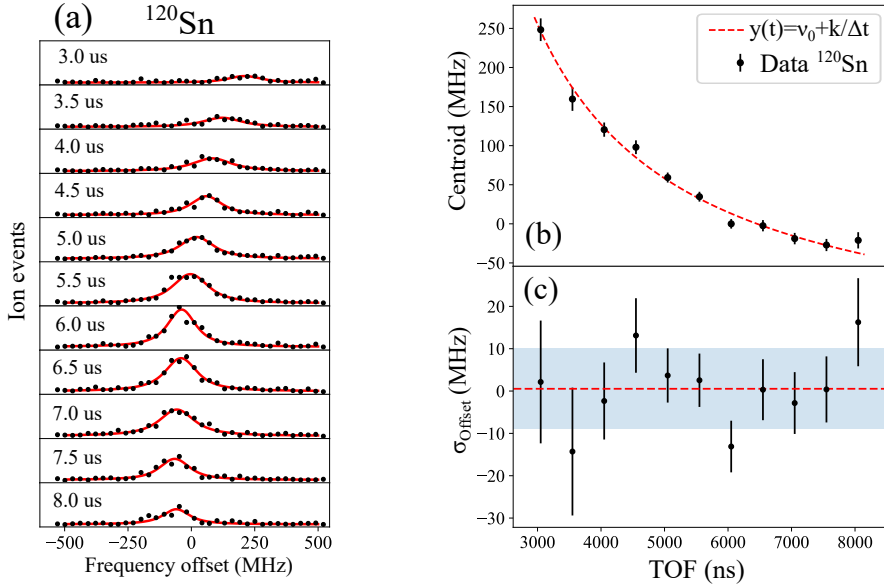


Figure 5.7: The spectrum is gated with different TOF and each gated (500 ns gate) spectrum fitted in (a). In (b) the trend in centroid frequency with TOF is fitted with the suggested function indicating excellent agreement.

Once the function $\nu(t)$ is determined the centroid frequencies with respect to TOF can be corrected accordingly $\nu_0 \approx \nu(t) - \frac{k}{\Delta t}$ thereby removing the frequency dependence on TOF. This correction procedure reduces the Doppler spread which results in a narrowing of the resonance peak, as demonstrated in the spectrum in Fig. 5.8. As seen in Tab. 5.1, the quality of the spectrum is improved with reduced background from the narrowing of the TOF gate together with a reduced linewidth by a factor of 2, resulting from the correction procedure. The centroid offset observed in Fig. 5.8 is due to the arbitrarily chosen reference frequency to which the data is corrected to. Since the entire spectrum is corrected equally using $\frac{k}{\Delta t}$ there is minimal influence to the relative peak locations after this correction procedure, as demonstrated in Ref. [107].

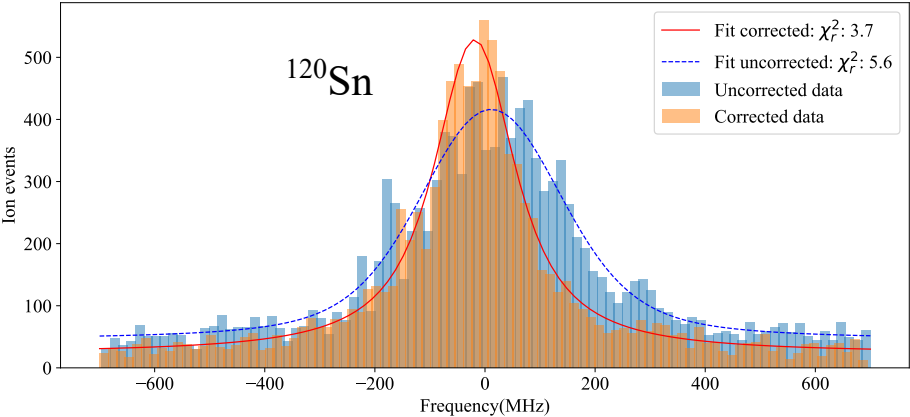


Figure 5.8: The difference between the uncorrected and corrected spectra using the TOF or 'banana'-correction procedure. The characteristics of the fitted voigt profiles are shown in Tab. 5.1.

Table 5.1: Difference between the fitted ^{120}Sn resonance peak before and after the 'banana'-correction procedure. The background is reduced due to the reduced TOF gate on the TOF bunch.

	Uncorrected		Corrected	
	Value	err	Value	err
χ_r^2	5.6		3.7	
FWHMT	312	6	178	8
FWHMG	236	20	54	20
FWHML	128	26	161	9
Background	45	3	24	2

Chapter 6

Tin ionization scheme development work

Transitions between atomic levels are unique for every element. The resonant steps required to step-wise excite and ionize the element therefore need to be carefully tailored in order to perform CRIS measurement of atomic tin which are sensitive to the nuclear structure observables. A non-exhaustive list of criteria need to be carefully considered when selecting resonant ionization schemes for performing a CRIS measurement:

- Accessibility - The transition need to be accessible using the available lasers at the CRIS setup. While lasers covering a large spectral bandwidth are available at CRIS, high resolution measurements required for resolving the hyperfine structures are limited to the Ti:Sa laser range and its higher harmonics.
- Ionization efficiency - The probed lower atomic state should have a high population after the charge-exchange neutralization process for maximizing the ionization efficiency.
- Transition strength and lifetime - The atomic transition strength should considered such that the laser power density is available for saturating the transitions used in the ionization scheme. Atomic states with longer lifetimes (and thus smaller natural linewidth) are preferred when performing precision measurements.

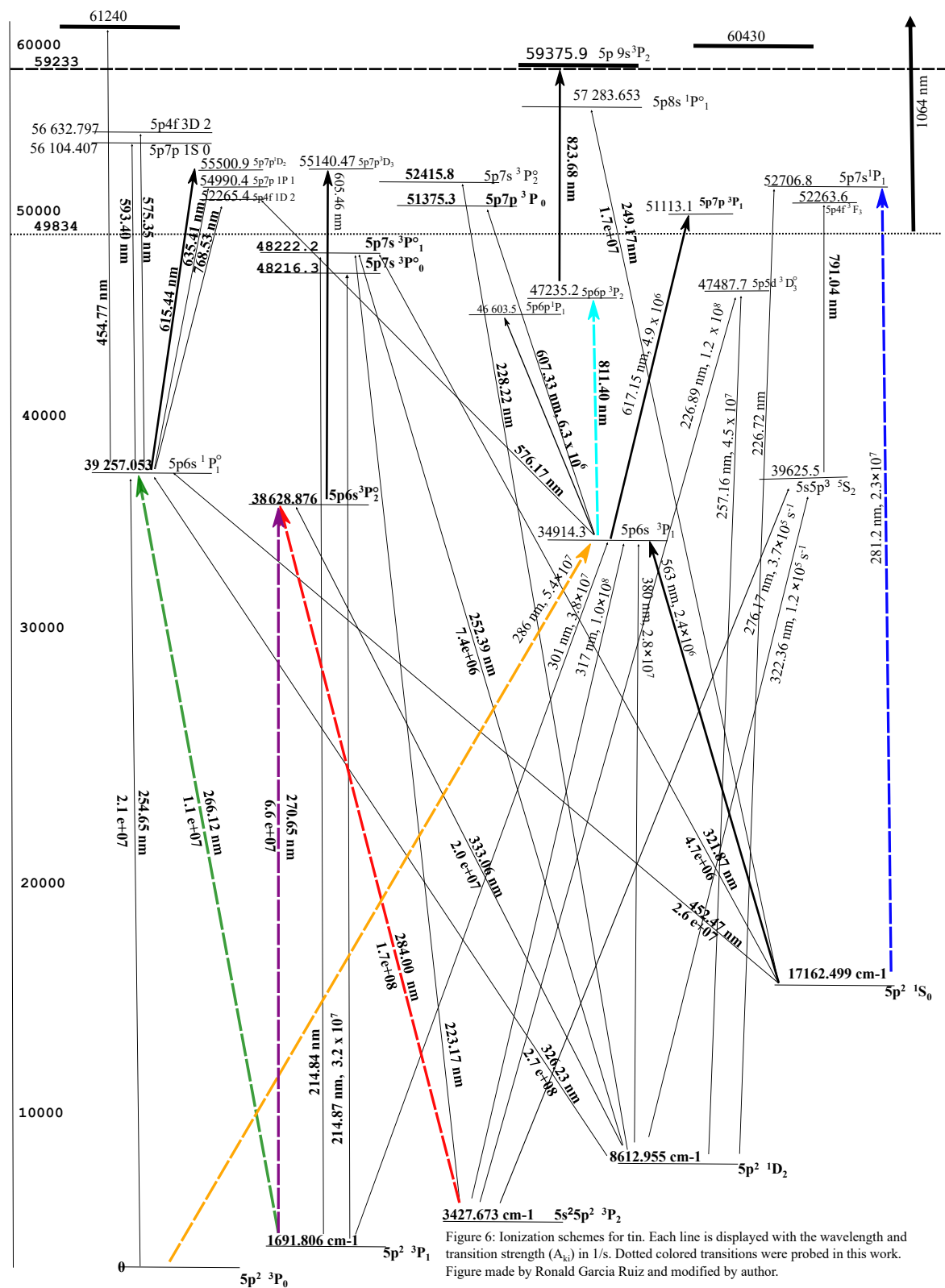
- Selectivity - Ionization schemes with multiple resonant steps might enhance isotope selectivity by reducing probability to resonantly excite isobaric contamination.
- Sensitivity to nuclear structure - The probed atomic state need to generate a considerable magnetic and electric field at the nucleus for accessing the nuclear moments. The transitions need to be chosen between atomic states which has considerable overlapping wavefunction with the nucleus for accessing its charge radius.

While spectroscopic measurements of tin have been performed for over a century [168], only a few resonance ionization schemes had been investigated prior to this work [137, 169, 170, 171]. Moreover, only a few atomic levels had a known sensitivity to the nuclear observables [172, 173, 174, 77]. Thus a comprehensive understanding of the above mentioned criteria was lacking, which motivated systematic measurements of laser ionization schemes used for probing the nuclear structure of the short-lived isotopes.

This chapter presents spectroscopic measurements of all ten stable isotopes produced using the laser ablation ion source (Sec. 4.3) for investigating the different resonance ionization schemes in tin based on the above mentioned criteria.

6.1 Ionization scheme considerations

The atomic electrons of tin occupy the ground-state in the $[\text{Kr}]4d^{10}5s^25p^2$ orbital configuration. The five lowest lying fine structure levels, namely $5p^2\ ^3P_{1,2,3}$, 1D_2 and 1S_0 , are expected to have a relative population between 5 to 30% after the neutralization charge-exchange-interaction with either Na or K, see Fig. 4.9 and 4.10. Atomic transitions considered for probing at CRIS with transition rates $A_{ki} > 10^6\text{ s}^{-1}$ were compiled from the NIST database [147], the ionization schemes are indicated in the scheme map in Fig. 6. The transition schemes probed in this work are presented in bold with the transitions probed in high resolution $< 1\text{ GHz}$ highlighted with colored dashed lines. The wavelength and transition strength are displayed for most transition lines. Non-resonant ionization pulsed 1064 nm light can be achieved from levels above 49834 cm^{-1} as indicated with a dotted line in the scheme map.



6.2 Paper I: Tin resonance-ionization schemes for atomic- and nuclear-structure studies

The following paper presents the extensive precision study of atomic levels in tin measured using the CRIS technique. Since the nuclear structure of the stable tin isotopes have been extensively studied in a wide range of experiments it allows us to decouple the nuclear structure contribution when studying the sensitivity of each atomic level to the nuclear observable. This paper presents the most comprehensive overview to date of tin atomic levels and their sensitivity to the magnetic dipole and electric quadrupole moment, in addition to the nuclear charge radii. This work combines theoretical calculations using a variety of modern theoretical approaches. This answers the question which ionization scheme is most sensitive to the nuclear observables of interest.

I was major contributor to the experimental work, I analyzed the experimental data and discussed the results with collaborators who performed the calculations while leading the writing of the manuscript.

Tin resonance-ionization schemes for atomic- and nuclear-structure studies

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This paper presents high-precision spectroscopic measurements of atomic tin using five different resonance-ionization schemes performed with the collinear resonance-ionization spectroscopy technique. Isotope shifts were measured for the stable tin isotopes from the $5s^25p^2\ ^3P_{0,1,2}$ and 1S_0 to the $5s^25p6s\ ^1P_1$, $^3P_{1,2}$ and $5s^25p7s\ ^1P_1$ atomic levels. The magnetic dipole hyperfine constants A_{hf} have been extracted for six atomic levels with electron angular momentum $J > 0$ from the hyperfine structures of nuclear spin $I = 1/2$ tin isotopes, $^{115,117,119}\text{Sn}$. State-of-the-art atomic calculations using a relativistic Fock-space coupled-cluster method and the configuration interaction approach combined with many-body perturbation theory allow accurate and reliable calculations of both field- and mass-shift factors for the studied transitions, in addition to the hyperfine magnetic fields and electric-field gradients of the atomic levels. The excellent agreement with the experimental results highlights the accuracy of modern atomic theory and establishes an important foundation for precision measurements of nuclear moments and charge radii of the most exotic isotopes of tin.

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I. INTRODUCTION

Atomic and ionic spectral lines are observed to shift between different isotopes of the same element. The isotope shift can be extracted using various optical methods, even for short-lived radioactive isotopes [1]. Precision measurements of isotope shifts, which are usually smaller than 1 part in 10^6 of the absolute transition frequency, provide insight into the electron-nucleus interaction and allow systematic studies of changes in nuclear charge radii [2,3] or probing of fundamental interactions [4–6]. This requires an accurate understanding of the atomic field- and mass-shift factors for decoupling the atomic-physics contribution [7,8]. Furthermore, the electronic fine-structure levels split into sublevels known as the

hyperfine structure (HFS) for atoms containing nuclei with nonzero spin. This splitting can be attributed to the presence of nonzero nuclear electromagnetic moments and depends on the hyperfine magnetic field and, for spin greater than $1/2$, on the electric-field gradient (EFG) generated by the electrons.

Until recently, field and mass shifts, hyperfine magnetic fields, and EFGs remained difficult to calculate with high accuracy for multielectron systems [7,9,10]. To calculate these properties, a sophisticated treatment of electron correlations and relativistic effects is required. Measurements of HFS constants of multielectron systems can, therefore, be of great value for validating modern electronic structure methods [11–14]. Furthermore, nuclear moments and charge radii, regularly extracted from the HFS and isotope shift measurements of stable and radioactive isotopes, are valuable for studies of nuclear structure [1–3,15–17]. Longstanding questions on the evolution of these nuclear structure observables towards the limits of existence test our description of the nuclear many-body problem.

Tin, having a closed shell for protons within the nuclear shell model ($Z = 50$), has the largest number of stable isotopes of all known elements, featuring seven even-mass spin-0 isotopes $^{112,114,116,118,120,122,124}\text{Sn}$, and three spin- $1/2$ isotopes $^{115,117,119}\text{Sn}$. For this reason, the tin isotopic chain provides

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an excellent case for investigating the influence of nuclear charge radii and nuclear magnetic dipole moments on the hyperfine structure of different atomic states. Moreover, tin is of particular interest for nuclear structure studies, as it is the heaviest known element containing two isotopes with both closed neutron and proton shells: the neutron-deficient ^{100}Sn and the neutron-rich ^{132}Sn [18–21]. Significant efforts have been made to perform laser spectroscopy across the tin chain reaching from ^{134}Sn down to ^{108}Sn [20,22–25]. Further laser-spectroscopy measurements are planned, both at existing facilities and in new laboratories currently under construction [26–28]. All proposed experimental methods aim to achieve high signal-detection efficiency and low background, a requirement to allow the study of exotic isotopes with low production rates when approaching ^{100}Sn . For example, at the ISOLDE radioactive beam facility of CERN, ^{101}Sn with a half-life of only 1.9(3) s [29] is predicted to be produced at a rate of less than one ion per second. A campaign for studying the neutron-deficient tin isotopes is ongoing at the Collinear Resonance Ionization Spectroscopy (CRIS) experiment at ISOLDE [26]. Other high-precision and sensitive laser-ionization-spectroscopy techniques are being developed, such as the perpendicularly illuminated laser ion source and trap (PI-LIST) method [30] and the in-gas-jet laser-spectroscopy method [31]. All these methods share a common need for well-understood laser-ionization schemes. These schemes have to be optimized specifically for the element of interest, with optimal sensitivity to the nuclear observables.

In this work, several laser-ionization schemes are evaluated, through measurements of isotope shifts and hyperfine-structure constants of all stable tin isotopes. Since there are no stable tin isotopes with nuclear spin greater than $I = 1/2$, there is no measurable electric quadrupole interaction between the nucleus and the surrounding electrons. Calculations of the EFGs are therefore required to evaluate the sensitivity of the atomic states to the spectroscopic nuclear electric quadrupole moment. The state-of-the-art relativistic Fock-space coupled-cluster (FSCC) approach was used for calculating the EFGs, hyperfine magnetic fields, and field-shift factors, while the configuration interaction approach with many-body perturbation theory (CI+MBPT) was used to calculate the mass-shift factors. The calculated atomic factors are validated by the experimental data and other measurements from the recent literature.

This paper is organized as follows. The CRIS technique used to perform the measurements is described in Sec. II, from the production of the ions of naturally abundant tin, to the collinear resonance ionization and ion detection. A detailed description of the laser setup used to generate the light required for laser ionization is further described, followed by a description of the data collection procedure. In Sec. III, the theoretical methods used in this work are introduced and the procedures used for calculating the field- and mass-shift factors, hyperfine magnetic fields, and EFGs are presented. Section IV introduces the data handling and analysis strategy that was followed to extract the atomic observables and their corresponding uncertainties. The hyperfine A_{hf} constants for all investigated atomic states in the odd-mass isotopes $^{115,117,119}\text{Sn}$ are presented and compared with the literature,

as are the isotope shifts for all transitions employed in this work. These results are discussed in Sec. V. In Sec. VA, the calculated field- and mass-shift factors are compared with experimental values deduced from a King-plot analysis against mean-square nuclear charge radii evaluated from a combined analysis of atomic spectroscopy, muonic x-ray, and electron-scattering data. Then, using the calculated field- and mass-shift factors, independent sets of root-mean-square (rms) nuclear charge radii for all stable isotopes of tin are deduced for two of the studied transitions and compared with the literature. Both approaches show that reliable field- and mass-shift factors are calculated. In Sec. VB, the hyperfine magnetic fields extracted from the data are compared with the theoretical predictions and experimental data from other work. This is followed by Sec. VC, where the calculated EFGs of the studied atomic levels are provided. The paper is concluded with a summary in Sec. VD.

II. EXPERIMENTAL SETUP

A. Ion source and the CRIS beam line

The ion source and the CRIS beam line [2,32,33] at ISOLDE-CERN are shown schematically in Fig. 1. A bunched ion beam containing all stable isotopes of tin was generated by laser ablation of a solid tin target (>99% purity). A 532 nm Nd:YAG laser was used for the ablation, operating at 100 Hz repetition rate and with approximately 30 mJ/pulse, focused onto the tin target, with a spot diameter less than 1 mm. The interaction with the laser pulse generated a plume of tin ions which was extracted by a low electric field. A detailed description of this unit is presented in Ref. [34]. The bunched beam of tin ions was accelerated with a voltage selected between 20 and 30 kV. By electrostatically steering the beam through a heated charge-exchange cell (CEC), which contains sodium vapor at 230–250 °C, the beam was partially neutralized. The fraction of neutral tin atoms within the beam varied between 10 and 30%, depending on the vapor pressure of the sodium. Different atomic states in Sn I were populated [35]. The partially neutralized beam leaving the CEC passed an electric-field gradient of 5000 V/cm perpendicular to its path, thereby removing ions and field-ionizing atoms neutralized into high-lying Rydberg states, before entering a differential pumping region held at a pressure of 5×10^{-9} mbar. Residual ions collisionally ionized in the differential pumping region were removed from the beam after passing an electric field of 1000 V/cm before entering the interaction region, which is kept at an ultrahigh vacuum of 5×10^{-10} mbar. Within the interaction region, each atom bunch was collinearly overlapped in space and synchronized in time with a series of laser pulses: first, a tunable high-resolution pulse which, when in resonance with the chosen atomic transition, excited the atomic electrons to a higher-lying atomic state. For the three-step ionization schemes, a second laser pulse resonantly excited the atoms into an even higher-lying state, before a third high-power laser pulse nonresonantly ionized the excited atoms. Performing this resonance-ionization process in flight reduces the Doppler broadening by a factor of 10^3 , often reaching the magnitude of the intrinsic width of the atomic transition [36]. Ions present in the bunch were then electrostatically deflected and counted

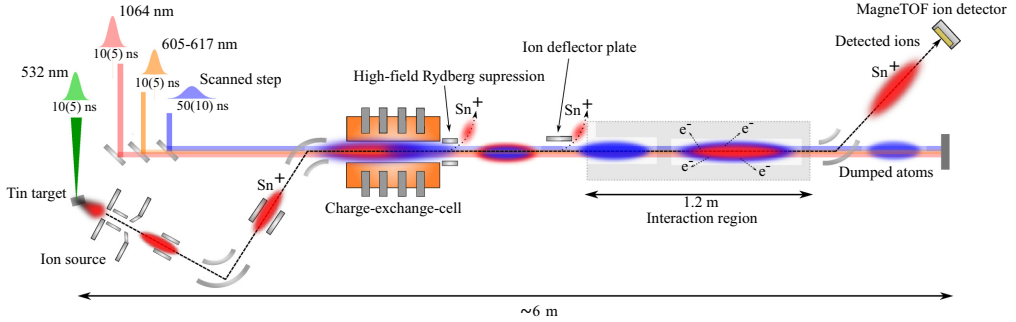


FIG. 1. Schematic overview of the CRIS beam line and the laser ablation ion source. The ions, indicated in red, are extracted from the solid tin target, accelerated, and steered through a charge-exchange cell. Here, the ions neutralize; only the atoms, indicated in blue, pass the high-field Rydberg suppression plates and the ion deflector before entering the interaction region. In the interaction region, laser pulses are collinearly overlapped with the atomic bunch in both time and space, resulting in stepwise excitation and ionization of the atoms when on resonance. The ions are separated from the remaining background atoms and detected with an ETP MagneTOF ion detector.

using an ETP DM291 MagneTOF detector. Any remaining atoms do not reach the detector, resulting in a low detected background signal.

B. Lasers

The tested laser-ionization schemes are shown in Fig. 2. The first laser step for all indicated ionization schemes was scanned in frequency in order to measure the hyperfine structures and isotope shifts. The narrowband laser pulses, required to resolve these effects, were generated from an injection-locked Ti:sapphire laser system [37] seeded with continuous-wave light produced by either an M-Squared SolisTiS or a Sirah Matisse Ti:sapphire laser system. This seeded laser is pumped using a Q-switched Nd:YAG laser (Lee Laser LDP-100MQG) at 1 kHz. In order to reach 266–286 nm, a

sum-harmonic generation unit, using two nonlinear crystals, i.e., bismuth borate (BIBO) and beta barium borate (BBO) crystals, was used. This provided laser pulses with a linewidth of the order of 60 MHz, a pulse length of 50(10) ns, and energy of 30–150 nJ/pulse. The laser frequency of the seed light was scanned at a rate below 5 MHz/s over the frequency region containing all stable tin isotopes and was continuously measured using a WSU-2 HighFinesse wave meter. This wave meter also recorded a reference wavelength from a stabilized laser diode (Toptica DLC DL PRO 780) locked to the Doppler-free hyperfine transition $5S_{1/2}(F=2) - 5P_{3/2}(F'=3)$ of ^{87}Rb (TEM CoSy). This was used to correct for drifts in the wave-meter readout over the course of the experiment [38]. In most cases, a second resonant step was used with a wavelength of 605–617 nm, a pulse energy of 50–200 μJ /pulse, and a pulse length of 10(5) ns. This laser light was produced with a SPECTRON SL4000 pulsed dye laser system, using DCM dye dissolved in ethanol. This dye laser was pumped using one of the 532 nm outputs from a dual-head Nd:YAG laser (Litron LPY 601 50-100 PIV), Q-switched at 100 Hz repetition rate. The second independent head provided pulsed laser light for laser ablation, thus defining the cycle length of measurements as 10 ms. The last nonresonant step is provided by a Nd:YAG laser (Litron TRLi 250-100) generating 50–100 mJ/pulse at 1064 nm with a pulse length of 10(5) ns and a repetition rate of 100 Hz. The delay and synchronization of all lasers was performed using a set of Quantum Composers 9528 digital-delay pulse generators. Due to the short half life of the first-excited states, which were all of the order of a few nanoseconds [39], all lasers were overlapped with a delay of 30(20) ns between subsequent pulses. Measurements of the full range of isotopes were repeated 2–3 times for each transition.

C. Data-collection procedure

The resonantly ionized ions were registered on an event-by-event basis, with sub-ns timing resolution for time-of-flight

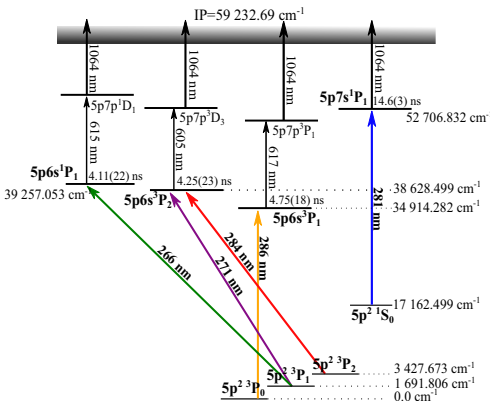


FIG. 2. Tin ionization schemes studied in this work. Lifetimes are taken from Ref. [39] and level energies from Ref. [41].

(TOF) determination from the pulsed laser-induced ablation production process to detection with an ETP MagneTOF detector, using a ChronoLogic TimeTagger4-2G time-to-digital card [40]. The frequency measurements of the first and second resonance steps including the reference diode were recorded every 10 ms in sync with the laser atom beam overlap. A time stamp for each measurement allowed the merging of frequency, TOF, and ion events which was binned with respect to frequency for generating a spectrum.

III. ATOMIC THEORY

A. Field-shift and HFS parameter calculations

High-accuracy calculations of the properties of heavy atoms require a high-quality treatment of relativistic and electron correlation effects. Therefore, a state-of-the-art relativistic Fock-space coupled-cluster approach and a combination of the configuration interaction with the many-body perturbation theory were employed for the investigation of the HFS and isotope shift parameters. The multireference relativistic FSCC method [42,43], employed in this work for the calculation of the field shifts, EFGs, and the hyperfine magnetic field constants, is considered to be one of the most powerful approaches for heavy many-electron systems. Compared to single-reference methods, it has the distinct advantage of being able to treat many excited states with consistent quality. The calculations are carried out in the framework of the Dirac-Coulomb Hamiltonian,

$$H_0 = \sum_i h_D(i) + \frac{1}{2} \sum_{i \neq j} \frac{1}{r_{ij}}, \quad (1)$$

where h_D is the Dirac one-particle Hamiltonian,

$$h_D(i) = c \boldsymbol{\alpha}_i \cdot \mathbf{p}_i + c^2(\beta_i - 1) + V_{\text{eN}}(i), \quad (2)$$

and α and β are the Dirac matrices. The Coulomb potential $V_{\text{eN}}(i)$ is the electron-nucleus interaction, with the nuclear charge distribution modeled by a Gaussian function as described in Ref. [44].

The FSCC method requires a closed-shell reference state from which the ground state and the excited states can be reached by adding or removing electrons. In the case of neutral tin, which has an open-shell ground-state electron configuration $[\text{Kr}]4d^{10}5s^25p^3$, the closed-shell Sn^{2+} system ($[\text{Kr}]4d^{10}5s^2$) is used as the reference state and two electrons are added to the orbitals that comprise the model space, P . By including the $5p$ orbital in P , the $5p^2$ configurations including the ground state of neutral tin are obtained. By allowing the electrons to occupy higher virtual orbitals, further excited states of interest can be included in the calculation. A larger model space also increases the accuracy because of the correlation contribution from the additional states. The use of large model spaces can, however, cause convergence difficulties due to the existence of close-lying states in the virtual space that couple strongly to the model-space states. The intermediate Hamiltonian (IH) approach [45] was applied to avoid this problem. In this method, the model space is divided into a main space P_m and an intermediate space P_i and the problematic amplitudes between the P_i and the virtual space are removed.

The field-shift factors F_λ are obtained by performing FSCC calculations of the transition energies for several values of $\langle r^2 \rangle$ by modifying the exponent of the Gaussian function describing the nuclear charge distribution [44]. The derivative of the energy with respect to $\langle r^2 \rangle$ then gives the field-shift factor for each transition.

The finite field method [46] was used for the calculation of the EFG (q) and the magnetic field at the core, A_0 , as described in Refs. [11,47]. The desired property is added to the Dirac-Coulomb Hamiltonian [Eq. (1)] as a perturbation H' with strength λ ,

$$H(\lambda) = H_0 + \lambda H'. \quad (3)$$

The total energy can then be expressed via the Taylor expansion,

$$E(\lambda) = E_0 + \lambda \left. \frac{dE_\lambda}{d\lambda} \right|_{\lambda=0} + \frac{\lambda^2}{2} \left. \frac{d^2E_\lambda}{d\lambda^2} \right|_{\lambda=0} + \dots \quad (4)$$

For small enough values of λ , higher-order terms can be ignored and the expectation value of H' can be obtained numerically, according to the Hellmann-Feynman theorem, from the first derivative of the energy with respect to λ ,

$$\left. \frac{dE(\lambda)}{d\lambda} \right|_{\lambda=0} = \langle \Psi | H' | \Psi \rangle. \quad (5)$$

The size of λ was chosen from two considerations: the perturbation must be large enough to avoid numerical errors, but small enough to stay in the linear regime where the higher-order terms can be neglected. The derivative is calculated from the two-point formula,

$$\left. \frac{dE_\lambda}{d\lambda} \right|_{\lambda=0} \approx \frac{E(\lambda) - E(-\lambda)}{2\lambda}. \quad (6)$$

In the case of the EFG calculation, H' is the interaction of the electric quadrupole moment Q with the EFG as given in Ref. [48]. Similarly for the calculation of A_0 , the magnetic hyperfine interaction is added as a perturbation; further details can be found in Ref. [11]. By using the two-point formula, any quadratic terms cancel out, resulting in an error proportional to λ^2 , as shown in Ref. [11]. Two versions of the DIRAC program package [49] were employed. The field-shift and EFG calculations were carried out using DIRAC15 [50], while DIRAC17 was used for the A_0 calculations [51]. The calculations were performed using the exact two-component Hamiltonian (X2C) [52]. This approach allows a significant decrease in computational time and expense, while reproducing very well the results obtained using the full four-component dc Hamiltonian, as tested for a variety of species and properties [53,54]. A comparison of the X2C and full four-component results for a small basis set and model space yielded differences of the order of 0.5%, thus justifying the use of the former approximation. The Gaunt term was included at the Dirac-Hartree-Fock step of the calculations using a small basis set and model space for uncertainty estimation purposes. It is part of the Breit interaction that corrects the two-electron part of the Dirac-Coulomb Hamiltonian up to the order of $(Z\alpha)^2$ [55].

An extensive investigation of the effect of various computational parameters (basis set, model space, etc.) on the

obtained results was carried out. The aim of this investigation was twofold. On the one hand, it allows one to identify the computational scheme where convergence of the obtained results is reached with respect to the various parameters, which is used to provide the final recommended values. On the other hand, this investigation is used to set uncertainties on our predictions.

The final recommended results are obtained using the relativistic core-valence 4-zeta basis set of Dyall [56] (cv4z), augmented by four diffuse functions in each symmetry in an even-tempered fashion. A large model space was used, consisting of the $5p\ 6s\ (5d\ 6p\ 7s\ 4f\ 6d\ 7p\ 8s\ 5f\ 7d\ 5g\ 8p\ 9s\ 6f\ 8d\ 9p\ 6g\ 6h\ 7g\ 10p\ 10s\ 7f)$ orbitals, where the orbitals in parentheses are in the intermediate space P_i . All the electrons were correlated and virtual orbitals with energies up to 500 a.u. were included in the calculation. For the calculation of F_A , a change in the radius (r^2) of approximately 20% was used, while the perturbation strength λ was 5×10^{-7} for the EFG calculations and 10^{-4} for A_0 . The remaining sources of error in these calculations are the basis-set incompleteness, neglect of higher-order relativistic effects (i.e., the full Breit and the QED corrections), and the residual correlation effects. The basis-set associated uncertainty is comprised of three parts to account for the basis-set quality, correlating functions, and augmentation. Basis-set quality was evaluated by comparing the results using a 3-zeta and the 4-zeta quality basis set. The effect of correlating functions is included by comparing the core-valence and all-electron basis-set results, while augmentation effects are included by comparing the results using a basis set augmented with three and with four diffuse functions in each symmetry. This adds up to 2% for all the values, except for the EFG and A_0 of the $5p7s\ ^1P_1$ and $5p6s\ ^1P_1$ states with uncertainties of about 15% and about 8%, respectively, and 15% for the field-shift factor of the $5p^2\ ^1S_0$ to $5p7s\ ^1P_1$ transition. It is assumed that the error due to the incomplete treatment of the relativistic effects is not larger than the difference between the X2C and the four-component results, together with the Gaunt contribution. It is about 1% for all states and transitions considered here. The missing correlation effects are due to the limited size of the correlation space and the finite size of the model space. The error caused by the limited correlation space is estimated by comparison of calculations with a virtual space cutoff at 500 and 2000 a.u., which gives an uncertainty of less than 0.1% for each property. The finite size of the model space is taken into account by comparing calculations that use a model space that differs by one orbital in each symmetry, resulting in an uncertainty of 1% for all the states except the highest one, where the effect is larger and of the order of 10%. In the present calculations, excitations beyond doubles are neglected. As triple and higher excitations cannot presently be evaluated for these properties within the FSCC framework, a rigorous scheme for estimating their size is not available. However, these corrections are usually quite small and are not expected to exceed about 3%, based on similar HFS calculations in Ref. [11]. Combining the above sources of error and assuming them to be independent, the total conservative uncertainty estimate on the calculated values is 4% for all the states and transitions except for the EFG and

A_0 of the $5p7s\ ^1P_1$ state, for which it is 18%, and the $5p6s\ ^1P_1$ state that has an uncertainty of 9%, and 15% for the field-shift factor of the $5p^2\ ^1S_0$ to $5p7s\ ^1P_1$ transition. The presented uncertainties are dominated by the basis-set and correlation effects.

B. Mass-shift calculations

To calculate the mass-shift factors, the combination of configuration interaction and many-body perturbation theory was used [57], implemented in the atomic structure code AMBIT (see [58] and references therein for full method and implementation details). These calculations start with Dirac-Fock equations to generate core orbitals and the V^{N-2} Dirac-Fock potential (which includes the electrostatic potential of core electrons from the closed-shell Sn^{2+} system). Valence and virtual orbitals were generated by diagonalizing a set of B splines over the Dirac-Fock operator [59].

A CI calculation was then performed including all single and double excitations from the reference configurations ($5p^2$, $5p\ 6s$, $5p\ 7s$, $5p\ 5d$) up to $16spdf$, which is close to saturation. In the CI+MBPT method, the two-electron Slater integrals are modified to include core-valence correlations to second order in the residual Coulomb interaction (formulas may be found in [60]). The MBPT basis includes all virtual orbitals up to $30spdfg$.

Despite making only a small contribution to the energy, configurations formed from excitations of the $5s^2$ “core” shell are found to have a large impact on the specific mass shift. Therefore, the $5s^2$ shell was removed from the MBPT expansion and included as a hole shell in the particle-hole CI+MBPT formalism [61]. Three-body MBPT diagrams [62] were included for any matrix elements involving the reference configurations listed above or the important $5s\ 5p^3$ configuration.

To calculate the relativistic mass shift, the finite field approximation was used with the relativistic formulation [63–65]

$$H'_{\text{MS}} = \frac{1}{2M} \sum_{i,j} \left\{ \mathbf{p}_i \cdot \mathbf{p}_j - \frac{Z\alpha}{r_i} \left[\boldsymbol{\alpha}_i + \frac{(\boldsymbol{\alpha}_i \cdot \mathbf{r}_i) \mathbf{r}_i}{r_i^2} \right] \cdot \mathbf{p}_j \right\}. \quad (7)$$

The mass shift can be separated into one-body and two-body operators, known as normal mass shift (NMS) and specific mass shift (SMS), respectively. In the Hamiltonian above, the normal mass shift is represented by terms with $i = j$ and the specific mass shift is given by terms $i \neq j$. The coefficient k_{MS} is defined from the expectation value of H'_{MS} using the finite field method [Eq. (5)].

To estimate the uncertainties, the calculation is repeated using a four-valence-electron approximation. While this is identical at the CI level to using the particle-hole formalism, the MBPT diagrams are changed considerably, in particular because subtraction diagrams are introduced [61]. This changes the calculated value of k_{SMS} considerably, although the NMS coefficients change by less than $\sim 10\%$. The difference between the four-valence-electron and two-valence-electron (particle-hole) calculation is used to estimate the theoretical uncertainty.

TABLE I. The atomic transitions studied in tin. The upper and lower energy levels are provided from Ref. [41], which are compared to the calculated FSCC values. The calculated values agree with the experimental data within less than 2%.

Transition	E_{lower} (cm ⁻¹)	E_{upper} (cm ⁻¹)	$E_{\text{FSCC}}^{\text{lower}}$ (cm ⁻¹)	$E_{\text{FSCC}}^{\text{upper}}$ (cm ⁻¹)
$5s^2 5p^2 \ ^3P_1 \rightarrow 5s^2 5p6s^1P_1$	1691.806	39257.053	1671	39124
$5s^2 5p^2 \ ^3P_1 \rightarrow 5s^2 5p6s^3P_2$	1691.806	38628.876	1671	38471
$5s^2 5p^2 \ ^1S_0 \rightarrow 5s^2 5p7s^1P_1$	17162.499	52706.832	17176	53331
$5s^2 5p^2 \ ^3P_2 \rightarrow 5s^2 5p6s^3P_2$	3427.673	38628.876	3361	38471
$5s^2 5p^2 \ ^3P_0 \rightarrow 5s^2 5p6s^3P_1$	0.000	34914.282	0	34767

IV. RESULTS

A. Atomic transitions and fine-structure levels

The transitions in Table I were studied using the laser-ionization schemes presented in Fig. 2. The calculated upper and lower level energies are compared to the experimental values in Table I. In the ground-state multiplet, the energy of the $5p^2 \ ^3P_1$ and $5p^2 \ ^3P_2$ levels is calculated with an accuracy of 1% and 2%, respectively, while for the excited states, an accuracy of less than 0.5% is achieved for all levels, except for the $5p6s \ ^3P_2$ level where it is 1%. Using these transitions, the spectra of $^{124-116}\text{Sn}$ shown in Fig. 3 were acquired with an acceleration potential of 20 kV. The $^3P_0 \rightarrow ^3P_1$ transition

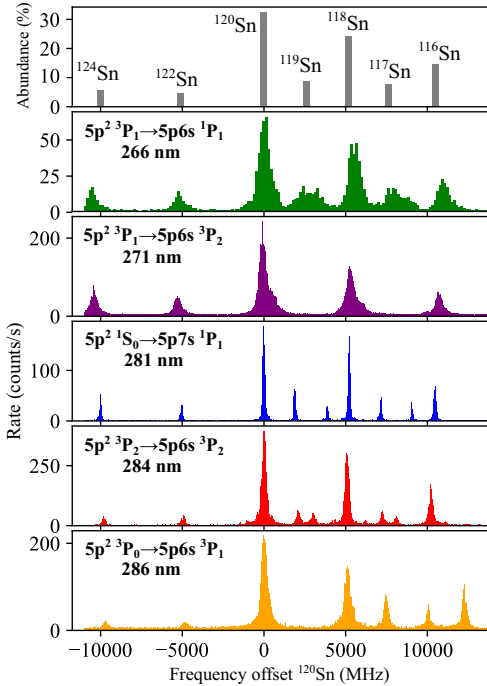


FIG. 3. Measured spectra of $^{116-124}\text{Sn}$ using laser-ionization schemes presented in Fig. 2. The abundances and center-of-mass location of the different isotopes are indicated on the top.

was also obtained using a 30 kV acceleration potential. That is because overlapping resonances were observed at the lower beam energy.

The peak shapes could be approximated with Voigt profiles with 200 to 600 MHz total FWHM, dominated by the Lorentzian contribution. Indeed, the upper state lifetimes, of the order of a few nanoseconds [39], were significantly smaller than the laser pulse length, thus requiring temporal overlap of the laser pulses. This resulted in power-broadening effects, induced by interactions with the high-power second and third laser steps [66]. Additionally, the velocity distribution of the ion beam causes an asymmetry of the peak profile, which is discussed further in Sec. IV C. Tin isotopes with a natural abundance higher than 4%, $^{116-124}\text{Sn}$ were observed with all ionization schemes. The isotopes $^{112,114,115}\text{Sn}$ with abundances below 1% could be resolved using the ionization schemes for the $^1S_0 \rightarrow ^1P_1$ and $^3P_2 \rightarrow ^3P_2$ transitions. These measurements indicate an abundance of 0.30(15)% of the least abundant stable isotope ^{115}Sn based on peak intensity ratios, in good agreement with the natural abundance in the literature [67]. For the three-step schemes, the dominant source of background was identified as nonresonant laser ionization due to the intermediate resonant step. This contribution accounted for roughly 80% of the observed background signal. This intermediate-step laser ionizes higher-lying atomic states populated during charge exchange, which were not field ionized before the interaction region. The remaining background was predominantly caused by collisional ionization after the interaction region, where a higher pressure of 10^{-8} mbar was present.

B. Isotope shifts and the hyperfine structure

The isotope shifts were determined from the difference in centroid frequencies ν^A in the rest frame of the atoms, using $\delta\nu^{A,A'} = \nu^A - \nu^{A'}$, where A' is the reference isotope chosen as ^{120}Sn due to its large relative abundance. For nonzero nuclear spin isotopes, the atomic fine-structure levels further break their degeneracy into a hyperfine structure, which can be calculated using the following expression:

$$\Delta E_{\text{hf}} = \frac{A_{\text{hf}}C}{2} + \frac{B_{\text{hf}}}{8} \frac{3C(C+1) - 4IJ(I+1)(J+1)}{IJ(2I-1)(2J-1)}, \quad (8)$$

with $C = F(F+1) - J(J+1) - I(I+1)$, and F takes integer steps between $|J-I| < F < J+I$, where I and J are the nuclear spin and electronic total angular momenta, respectively. The first dipole term is proportional to $A_{\text{hf}} = gA_0$, where A_0 is the magnetic field contribution at the core

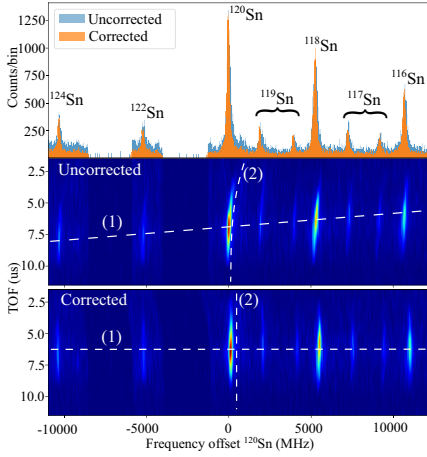


FIG. 4. Demonstration of TOF vs frequency correction procedure for the $^1S_0 \rightarrow ^1P_1$ transition. The applied correction procedure involves two steps: (1) the resonances of all isotopes are shifted to have the same median TOF and (2) the bananalike shape for each resonance is corrected according to the curvature of the reference isotope ^{120}Sn , reducing the linewidth and asymmetry of the final spectrum. The uncorrected and corrected histograms are projected at the top of the figure using 25 MHz bin size.

generated by the electrons, and $g = \mu_I/I$ is the ratio of the nuclear magnetic dipole moment μ_I to the nuclear spin I . The second term in Eq. (8) accounts for the electric quadrupole contribution and is proportional to $B_{\text{hf}} = qQ_s$, where q is the EFG generated by the electrons and Q_s is the spectroscopic electric quadrupole moment of the nucleus. This term is nonzero for $J, I > 1/2$. As the stable odd-mass isotopes, $^{115}\text{Sn}, ^{117}\text{Sn}$, have a ground-state nuclear spin $I = 1/2$, no quadrupole interactions can be studied with stable tin isotopes, and therefore the second term does not play a role in the analysis of the data.

C. Data-analysis procedure

Using the two-dimensional (2D) histogram of the TOF versus laser frequency, shown in Fig. 4, two corrections are applied. Initially, by gating on laser frequencies corresponding to the resonance signal for a specific tin isotope, TOF histograms for the different isotopes can be obtained. TOF histograms have a width of about $5(1) \mu\text{s}$. The median of the TOF histogram for each isotope is observed to exhibit a mass dependence, linear in first order, with an average slope of $150(50) \text{ ns/u}$ at a beam energy of $20\,000(50) \text{ eV}$. These TOF histograms for each selected mass were shifted to match the median of the TOF for the reference isotope ^{120}Sn ; see step (1) in Fig. 4. Shifting all masses to the same TOF allowed the use of a single TOF gate for all isotopes, conveniently reducing unwanted background without losing statistics. In a second step, as demonstrated in Ref. [34], the bananalike shape observed in the uncorrected TOF histogram in Fig. 4 for each of

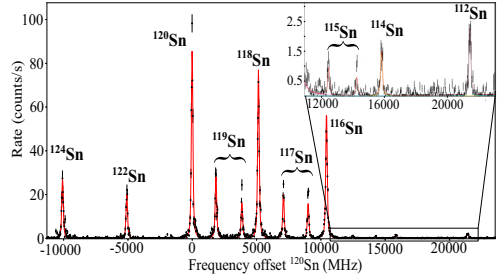


FIG. 5. A sample spectrum of the $^1S_0 \rightarrow ^1P_1$ transition. All stable isotopes can clearly be identified, including ^{115}Sn with a measured abundance of $0.30(15)\%$.

the isotope resonances is corrected. This shape is induced by an asymmetric ion velocity distribution, with a high velocity component from inhomogeneities in the extraction potential at the ion source and plasma-related effects from the ablation process [34]. Thus, ions with a different TOF experience a different Doppler shift, which effectively increases the linewidth in the laser scan and generates a tail extending towards higher frequencies. In order to correct for this energy spread, the data were subdivided in different TOF slices, 200–500 ns in width. The frequency spectrum obtained for each of these slices was then fitted with a Voigt profile using a χ^2 -minimization fitting routine to obtain the peak centroid. The centroids for each TOF slice show a clear correlation with the TOF, which is well approximated by the function $f(t) = k/t + c$. In this function, $f(t)$ is a frequency offset with respect to the TOF t , while k and c are free fit parameters. The frequency shift with respect to TOF for ^{120}Sn was used to correct the spectrum including all isotopes. The result is shown in Fig. 4, correction (2), leading to straight lines in the corrected TOF versus frequency plot. After the correction procedure, the total linewidth in the hyperfine spectrum is reduced and the background is suppressed as presented in the spectrum on top of Fig. 4. As an example, for the $^1S_0 \rightarrow ^1P_1$ transition, the total FWHM decreased from $253(5)$ to $173(4) \text{ MHz}$. The significance of this correction depends on the experimental conditions of the ion source. This correction procedure could be applied to the spectra obtained with the $^1S_0 \rightarrow ^1P_1$, $^3P_2 \rightarrow ^3P_2$, and $^3P_0 \rightarrow ^3P_1$ transitions, where accurate TOF data were available.

The corrected spectra were then fitted with a Voigt profile using χ^2 minimization, enabling extraction of magnetic hyperfine constants and centroid frequencies using the SATLAS analysis package [68]. A fitted spectrum of the $^1S_0 \rightarrow ^1P_1$ transition can be seen in Fig. 5. Each spectrum was fitted with various bin sizes in order to determine the dependence of the extracted parameters on the chosen bin size. The uncertainty associated with the TOF correction procedure is determined from the centroid scattering around the weighted mean of all fitted TOF slices. The combination of these contributions is listed in Table II as “Data processing unc.” since they are solely due to the data-handling procedure. Table II lists other uncertainties as well. The dominant source of uncertainty for the isotope shifts was associated with the ion beam energy, with an estimated stability of 0.001% of the set acceleration

TABLE II. Main sources of systematic errors identified from the extraction of isotopic shift and the hyperfine A_{hf} value for the 3P_2 state of ^{117}Sn using the $^3P_2 \rightarrow ^3P_2$ transition.

	A_{hf}^{117} (MHz)	$\delta\nu^{120,117}$ (MHz)
Value	-1546.0	-463.1
Data processing unc.	7.5	7.3
Ion beam energy	3.2	10.5
Wave-meter calib.	0.6	2.8
Total systematic unc.	8.2	13.1

voltage, and was furthermore only known with an accuracy of ± 50 V. The uncertainty due to laser frequency calibration, and possible nonlinearities from the wave-meter readout, are listed as the “wave-meter calib.” uncertainty.

The extracted A_{hf} constants for the different atomic states, seen in Table III, are compared with the literature values [22,23,69]. For ^{115}Sn , A_{hf} of the $5p^2\ ^3P_2$ state was fixed to a high-precision measurement from Ref. [69] during fitting of the data, obtained with the $^3P_2 \rightarrow ^3P_2$ transition, required as a constraint due to the low statistics. Similarly, for ^{119}Sn and ^{117}Sn , A_{hf} of the $5p^2\ ^3P_1$ state was fixed for the $^3P_1 \rightarrow ^1P_1$ transition. Additionally, the ratio of A_{hf} for $^{117,119}\text{Sn}$ was fixed to the same ratio as their nuclear magnetic moments from Ref. [25] for the $5p6s\ ^1P_1$ state. The A_{hf} constants are in good agreement with the literature within 1σ of the total uncertainty for all levels. Additionally, the isotope shifts using ^{120}Sn as reference are presented in Table IV. The isotope shifts of the $^3P_0 \rightarrow ^3P_1$ transition are compared with data from Ref. [22], indicating a close agreement with the literature.

V. DISCUSSION

A. Evaluating field- and mass-shift factors

The isotope shift is well approximated by the summed contribution from the field- and mass-shift terms accordingly,

$$\delta\nu^{A,A'} = F_\lambda \delta\langle r^2 \rangle^{A,A'} + k_{\text{MS}} \mu^{A,A'}. \quad (9)$$

Here, $\delta\langle r^2 \rangle^{A,A'}$ is the difference in mean-squared nuclear charge radii and $\mu^{A,A'} = \frac{m_A - m_{A'}}{m_A m_{A'}}$ is the reduced mass factor, where m is the atomic mass. The field-shift factor F_λ corresponds to the change of electronic charge density at the site of the nucleus, and thus determines the sensitivity to the change in the nuclear charge radius. The mass-shift factor k_{MS} is the sum of the normal (k_{NMS}) and specific (k_{SMS}) mass shifts. In order to extract F_λ and k_{MS} for each transition, a King-plot

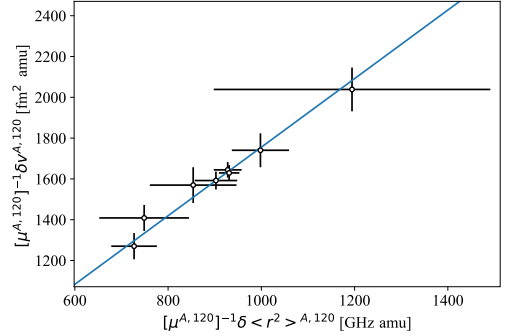


FIG. 6. King plot using Eq. (10) of the $^1S_0 \rightarrow ^1P_1$ transition with ^{120}Sn as a reference.

method as described in Ref. [70] was used, where the inverse of the reduced mass factor is multiplied on both sides of Eq. (9),

$$[\mu^{A,A'}]^{-1} \delta\nu^{A,A'} = F_\lambda [\mu^{A,A'}]^{-1} \delta\langle r^2 \rangle^{A,A'} + k_{\text{MS}}, \quad (10)$$

revealing a linear trend with slope F_λ and intercept k_{MS} . The isotope shifts in Table IV for each transition are compared with mean-squared nuclear charge radii determined from the combined analysis of muonic x-ray, electron-scattering, and optical data provided in Ref. [71] with a reduced mass factor calculated using masses from Ref. [72]. The King plot for each transition was fitted using orthogonal distance regression, which provides F_λ and k_{MS} from the slope and intercept. A sample King plot for the $^1S_0 \rightarrow ^1P_1$ transition is shown in Fig. 6. Separate King plots were created using $^{116,118,120}\text{Sn}$ as reference isotopes for each transition and the average F_λ and k_{MS} were selected for the final experimental values, with uncertainties determined from the standard deviation of the scatter summed in quadrature with the average sample error. Due to the lower statistics, the odd-mass isotopes were excluded from the King-plot analysis of the $^3P_1 \rightarrow ^1P_1$ transition.

F_λ and k_{MS} determined from experiment are compared to those from the atomic calculations, as described in Sec. III, and to the previous literature values for the $^3P_0 \rightarrow ^3P_1$ transition in Table V. Most transitions have similar values of F_λ , thus all being similarly sensitive to a change in the nuclear charge radius. Only the $^1S_0 \rightarrow ^1P_1$ transition has a

TABLE III. Hyperfine A_{hf} constants (in MHz) for $^{115,117,119}\text{Sn}$, compared with the literature (^aRef. [69], ^bRef. [22], and ^cRef. [23]). Statistical uncertainties are given in parentheses and experimental systematic uncertainties are given with square brackets.

Level	A_{Exp}^{115}	A_{Lit}^{115}	A_{Exp}^{117}	A_{Lit}^{117}	A_{Exp}^{119}	A_{Lit}^{119}
$5p^2\ ^3P_1$		507.445(4) ^a	695(83) [130]	552.608(4) ^a	452(82) [85]	578.296(4) ^a
$5p^2\ ^3P_2$		-1113.770 (4) ^a	-1211(9) [8]	-1212.956(3) ^a	-1269(6) [7]	-1269.419(3) ^a
$5p6s\ ^3P_1$		-4394.16(14) ^b	-4782(9) [22]	-4785.45(17) ^b	-5026(7) [25]	-5007.95(17) ^b
$5p6s\ ^3P_2$	-1438(9) [16]		-1546(9) [8]		-1621(6) [7]	
$5p6s\ ^1P_1$		-225.7 (9) ^c	-254(28) [23]	-247.6 (2) ^c	-266(29) [24]	-260.2 (2) ^c
$5p7s\ ^1P_1$	-1185(8) [13]		-1269(3) [12]		-1336(3) [11]	

TABLE IV. Measured isotope shifts (in MHz). Statistical uncertainties are indicated with parentheses and experimental systematic uncertainties are indicated with square brackets. For the $^3P_0 \rightarrow ^3P_1$ transition, a comparison is made to the literature [22].

Isotope	$\delta \nu^{A,120}_{^1S_0 \rightarrow ^1P_1}$	$\delta \nu^{A,120}_{^3P_2 \rightarrow ^3P_2}$	$\delta \nu^{A,120}_{^3P_1 \rightarrow ^3P_2}$	$\delta \nu^{A,120}_{^3P_1 \rightarrow ^1P_1}$	$\delta \nu^{A,120}_{^3P_0 \rightarrow ^3P_1}$	$\delta \nu^{A,120}_{^3P_0 \rightarrow ^3P_1}$ [22]
124	342(3) [17]	462(10) [15]	415(8) [29]	479(28) [15]	449(19) [20]	441.15(15)
122	192(3) [8]	228(11) [10]	214(9) [34]	238(29) [14]	270(15) [26]	235.35(15)
120	0	0	0	0	0	0
119	-142(2) [7]	-175(6) [8]	-166(17) [28]	-176(68) [93]	-211(7) [18]	-179.59(12)
118	-222(2) [12]	-267(4) [10]	-273(8) [33]	-263(17) [13]	-263(7) [18]	-270.24(15)
117	-372(3) [18]	-463(5) [13]	-467(41) [96]	-649(81) [23]	-494(9) [19]	-471.43(12)
116	-458(2) [12]	-580(6) [16]	-577(17) [26]	-622(15) [33]	-603(14) [39]	-576.04(15)
115	-637(5) [19]	-828(15) [19]				-804.92(12)
114	-722(5) [16]	-911(8) [23]				-900.68(15)
112	-971(5) [22]	-1253(6) [29]				-1218.29(15)

reduced F_λ by roughly 30% compared to the other transitions. Multiple independent studies have been performed using the $^3P_0 \rightarrow ^3P_1$ transition with varying estimates for the field-shift factor, i.e., 2390(270) [23], 3300(270) [24], 2180(170) [71], and 2740(570) MHz/fm² [20], and the mass-shift factor, i.e., -761(200) [24], 83(147) [71], and -455(479) GHz amu [20]. The weighted mean of these values for the $^3P_0 \rightarrow ^3P_1$ transition, with weights $1/\sigma^2$ and errors scaled with the reduced χ^2 , is presented in Table V. Both the experimental value and the calculation are in close agreement with the literature for both F_λ and k_{MS} for this transition. However, the calculations indicate a better agreement with the lower bound of the literature F_λ and upper bound of the literature k_{MS} , which are closer to the values determined in Ref. [71]. The overall agreement between the calculated and experimental values for each transition is very good, demonstrating that modern relativistic approaches are capable of producing reliable field- and mass-shift factors for many-electron atoms such as tin.

Using the measured isotope shifts given in Table IV, combined with the calculated F_λ^{FSCC} and $k_{MS}^{\text{CI+MBPT}}$ for each transition, independent sets of mean-squared nuclear charge radii can be deduced accordingly,

$$\delta \langle r^2 \rangle^{A,A'} = \frac{1}{F_\lambda^{\text{FSCC}}} (\delta \nu^{A,A'} - \mu^{A,A'} k_{MS}^{\text{CI+MBPT}}). \quad (11)$$

The rms charge radii $R_{\text{rms}} = \sqrt{\langle r^2 \rangle^{A,A'}}$ of the stable tin isotopes are determined using ^{120}Sn as a reference: $\langle r^2 \rangle^{1/2} = 4.654(1)$ fm [71]. As an example, the resulting radii determined using isotope shifts from the $^3P_2 \rightarrow ^3P_2$ and $^1S_0 \rightarrow ^1P_1$ transitions are displayed in Fig. 7 and compared

with radii from a combined analysis provided in Ref. [71]. This shows the good agreement with literature with only a slight deviation, well within the theoretical uncertainties. The high precision of the data reveals the small odd-even staggering effect displayed in the inset of Fig. 7, which is in excellent agreement with the literature and can provide an interesting insight into the nuclear structure [2].

B. Hyperfine magnetic fields

The hyperfine magnetic field contribution to A_{hf} can be decoupled from nuclear-structure effects by dividing with μ_I/I : $A_0 = A_{\text{hf}} I / \mu_I$. The A_0 value for each transition was extracted from the experimental A_{hf} of the three spin-1/2 isotopes, using magnetic dipole moments provided in Ref. [25]. The weighted means of A_0 from the three isotopes is presented in Table VI for each studied atomic level. All values are in close agreement with those from the literature, as well as with the calculated values from FSCC theory, visualized in Fig. 8(a). Atomic states with a larger magnetic hyperfine field are more sensitive for the measurement of nuclear magnetic moments. The hyperfine magnetic field varies significantly among the atomic configurations, with the $5p6s\ ^3P_1$ state exhibiting the greatest sensitivity.

C. Electric-field gradients

The EFGs calculated using the relativistic FSCC method are presented in Table VII and can be seen in Fig. 8(b). Confidence in the accuracy of the calculated EFGs is provided by the good agreement of F_λ and A_{hf} with the experimental results. Additionally, the EFGs of the $5p6s\ ^1P_1$ and $5p6s\ ^3P_1$

TABLE V. Calculated and experimental field- and mass-shift factors. The weighted mean of the experimentally determined field-shift factors from Refs. [20,23,24,71] and mass-shift factors from Refs. [20,24,71] for the $^3P_0 \rightarrow ^3P_1$ transition are shown in bold.

Transition	Field shift (MHz/fm ²)			Mass shift (GHz amu)				
	F_λ^{FSCC}	F_λ^{Exp}	F_λ^{Lit}	$k_{MS}^{\text{CI+MBPT NMS}}$	$k_{MS}^{\text{CI+MBPT SMS}}$	$k_{MS}^{\text{CI+MBPT MS}}$	k_{MS}^{Exp}	k_{MS}^{Lit}
$^1S_0 \rightarrow ^1P_1$	1552(233)	1584(209)		519	-409	110(300)	160(184)	
$^3P_2 \rightarrow ^3P_2$	2217(89)	2024(184)		604	-427	177(300)	205(160)	
$^3P_1 \rightarrow ^3P_2$	2200(88)	2323(395)		630	-436	193(300)	-116(328)	
$^3P_1 \rightarrow ^1P_1$	2104(84)	2932(1083)		645	-472	174(300)	-460(890)	
$^3P_0 \rightarrow ^3P_1$	2202(88)	2831(546)	2452(240)	598	-431	167(300)	-424(455)	-160(312)

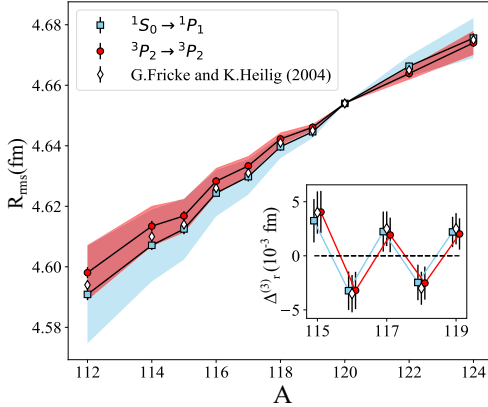


FIG. 7. The rms charge radii determined from the $^3P_2 \rightarrow ^3P_2$ and $^1S_0 \rightarrow ^1P_1$ transitions using Eq. (11) compared to the literature [71]. The total experimental uncertainty is indicated with error bars, and the theoretical uncertainty from $k_{\text{MS}}^{\text{CI+MBPT}}$ and $F_{\lambda}^{\text{FSCC}}$ is represented with color-coded bands. The inset displays the three-point odd-even staggering parameter $\Delta_r^{(3)} = \frac{1}{2}(R_{A+1} - 2R_A + R_{A-1})$ highlighting the small odd-even staggering effect that becomes visible thanks to the high precision of the data.

states were calculated in Ref. [23] and, more recently, using a wide variety of advanced computational approaches in Refs. [25,73]. All of them are in good agreement with the values calculated in this work. An accurate understanding of EFGs is essential for the determination of the electric quadrupole moments. From this work, the $5p6s\ ^3P_2$ state is identified to have the highest sensitivity to Q_s .

D. Summary

Isotope shifts and hyperfine magnetic fields have been extracted using five atomic transitions in tin. State-of-the-art relativistic FSCC calculations provide the isotope-independent hyperfine magnetic field parameters of the atomic levels, showing excellent agreement with experiment. Additionally, these calculations provide EFGs, which are not accessible through atomic spectroscopy measurements of the stable tin isotopes and are essential for studying the electric quadrupole

TABLE VI. A_0 values (in MHz) from this work compared with FSCC calculations and the literature values deduced from Refs. [22,23,69] in Table III. The A_0 values are determined using magnetic moments from Ref. [25] as $A_0 = A_{\text{hf}} I / \mu_I$.

Level	A_0^{Lit}	A_0^{Exp}	A_0^{FSCC}
$5p^2\ ^3P_1$	-276.77(2)	-278(29) [34]	-257(10)
$5p^2\ ^3P_2$	607.50(4)	607(2) [3]	598(24)
$5p6s\ ^3P_1$	2396.71(15)	2402(3) [11]	2352(94)
$5p6s\ ^3P_2$		777(2) [5]	783(31)
$5p6s\ ^1P_1$	124.3(2)	127(10) [16]	145(13)
$5p7s\ ^1P_1$		638(1) [8]	571(103)

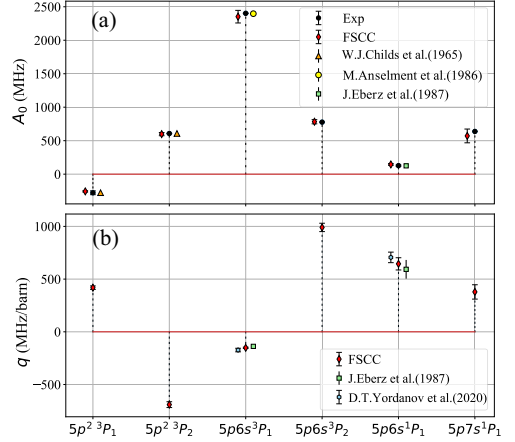


FIG. 8. (a) The A_0 values are compared with theoretical predictions using the FSCC method and the literature data from Refs. [22,23,69]. (b) The calculated EFGs are displayed and compared with other calculated values from Refs. [23,25]. The dotted lines represent the sensitivity to μ_I or Q_s , respectively. Only statistical errors are displayed for the experimental data from this work.

moment of the exotic tin nuclei using laser spectroscopy. According to these calculations, the $5p6s\ ^3P_2$ atomic level has the greatest sensitivity to the spectroscopic electric quadrupole moment, with an EFG roughly 50% greater compared to the $5p6s\ ^1P_1$ level used in previous work. The $5p6s\ ^3P_1$ level exhibits the greatest sensitivity to the nuclear magnetic moment, motivating its frequent use for probing the nuclear magnetic dipole moment. Furthermore, the large set of isotope shift measurements has been used for benchmarking the electronic F_{λ} and k_{MS} factors calculated using the combination of relativistic FSCC and CI+MBPT calculations. These calculations are in good agreement with experimentally obtained F_{λ} and k_{MS} factors of all studied transitions, allowing accurate determination of mean-square charge radii from isotope shift measurements.

In conclusion, this work demonstrates the reliability of modern atomic theory and provides a useful foundation for future measurements of the most short-lived exotic isotopes of tin from the evaluated sensitivity of several transitions to nuclear moments and mean-square nuclear charge radii.

TABLE VII. The EFGs (in MHz/b) calculated using FSCC compared with calculations from Refs. [23,25] with an estimated uncertainty of 15% for the values provided in Ref. [23].

Level	$q_{\text{Eberz}} [23]$	$q_{\text{MCDHF}} [25]$	q_{FSCC}
$5p^2\ ^3P_1$			419(17)
$5p^2\ ^3P_2$			-691(28)
$5p6s\ ^3P_1$	-138(21)	-173(17)	-152(6)
$5p6s\ ^3P_2$			990(40)
$5p6s\ ^1P_1$	593(89)	706(50)	645(58)
$5p7s\ ^1P_1$			378(68)

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6.3 The natural abundances of stable tin isotopes

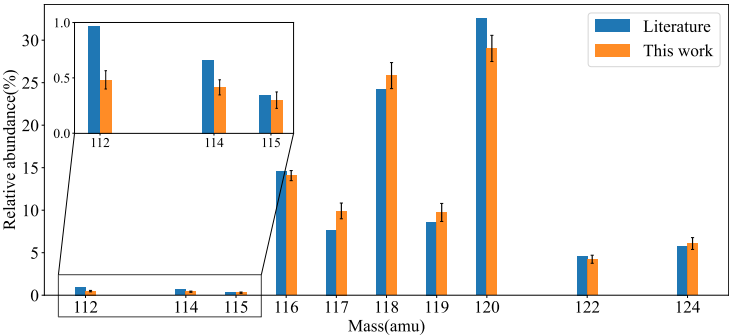


Figure 6.1: Tin isotope abundances measured using the CRIS technique compared with the natural abundances.

All ten stable tin isotopes could be resolved using the $5p^2\ ^1S_0 \xrightarrow{281\text{nm}} 5p7s\ ^1P_1 \xrightarrow{1064\text{nm}} IP$ transition scheme. The integrated intensity of the fitted spectra of all stable tin isotopes were normalized to provide an estimate of the isotopic abundances. The determined abundances from the CRIS measurement are presented in Fig. 6.1 and in Tab. 6.1 together with the natural abundances [175]. The isotopic abundances of the tin sample used in the laser ablation ion source are in good agreement with the abundances of natural tin except for the lightest tin isotopes. Measurements were performed by scanning from ^{124}Sn down to ^{112}Sn . The slow decay in ion yield from the ablation ion source over the duration of the measurements (4h) is a likely reason for this offset.

Table 6.1: Measured abundances of stable tin isotopes using CRIS.

Isotope	Natural abundance(%)	Measured abundance(%)	error(%)
124	5.79	6.08	0.70
122	4.63	4.23	0.47
120	32.58	29.03	1.54
119	8.59	9.74	1.06
118	24.22	25.83	1.53
117	7.68	9.92	0.93
116	14.54	14.07	0.59
115	0.34	0.30	0.07
114	0.66	0.41	0.07
112	0.97	0.48	0.08

6.4 Scanning the $5p6s^1P_1 \rightarrow 5p6p^3P_2$ transition

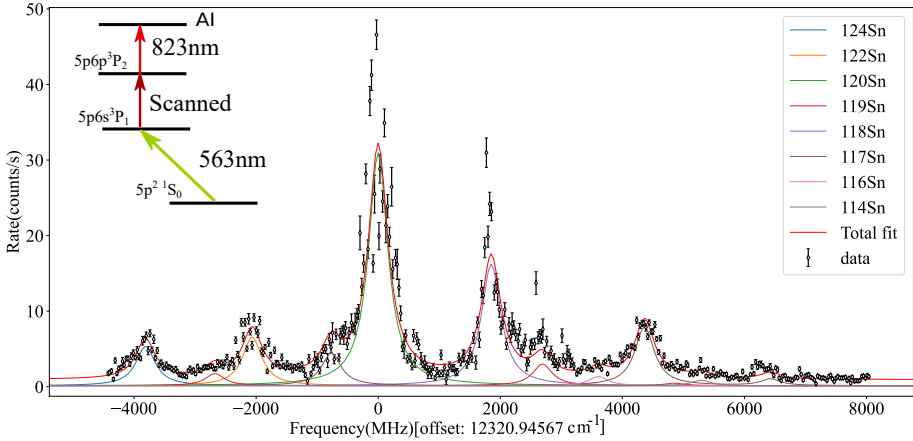


Figure 6.2: Observed tin resonances from scanning $5p6s^1P_1 \rightarrow 5p6p^3P_2$ as an intermediate step in a resonance scheme presented in the inset diagram.

Beyond the work published in Ref. [107], an ionization scheme to the $5p7p^3P_2$ auto-ionizing (AI) state was explored. In this scheme the $5p6s^1P_1$ state was optically pumped from the $5p^2^1S_0$ meta-stable state, using 563 nm light from the PDL with 300 $\mu\text{J}/\text{pulse}$. Fundamental light from the injection-seeded Ti:Sa laser was used to probe the intermediate transition $5p6s^1P_1 \rightarrow 5p6p^3P_2$ with 6 $\mu\text{J}/\text{pulse}$, a final resonant laser step at 823 nm from the Z-cavity Ti:Sa was used to excite into the $5p7p^3P_2$ auto-ionizing state with 34 $\mu\text{J}/\text{pulse}$. The resulting measurement is shown in Fig. 6.2.

While the spin-0 isotopes could be resolved, the hyperfine splittings of the spin-1/2 isotopes $^{119,117}\text{Sn}$ were difficult to extract due to the low statistics with three expected peaks for a $J = 1 \rightarrow J' = 2$ transition. Suggested fits of the hyperfine structures were achieved by fixing the A_{hf} -values for the $5p6s^1P_1$ state, by locking the A_{hf} -ratio using magnetic moments from [77] and fixing the hyperfine peak intensities according to the Racah intensities, Eqn. 3.26.

With these things considered the hyperfine structures were fitted with the A_{hf} -values provided in Tab. 6.2. Using g-factors from D.T Yordanov *et al.* [77], the hyperfine magnetic field for the $5p6p^3P_2$ atomic state could be determined to $A_0 = 414(6)[22]$ MHz. This is about 30% greater than the hyperfine magnetic field of the $5p6p^1P_1$ state measured by S. G. Nakhate *et al.* [171]. Furthermore,

FSCC calculations of the state not included in paper I is in reasonable agreement with this value assuming a 15% uncertainty as shown in Tab. 6.2.

Despite the enhanced cross-section by ionizing into an AI-state only low statistics measurements could be achieved using this scheme, due to significant non-resonant background associated with the 823 nm light. Non-resonant ionization using high-power 1064 nm pulsed light was therefore used as it was sufficient to saturate the ionization process.

Table 6.2: Measured A_{hf} -values for the $5p6p^3P_2$ atomic state. The statistical uncertainties are presented in round brackets with systematical uncertainties in square brackets. A 15% uncertainty is estimated for the FSCC calculated values.

Isotope	A_{hf}^{Exp} [MHz]	A_0^{Exp} [MHz]	A_0^{FSCC} [MHz]
119	-865(12)[45]	414(6)[22]	349[52]
117	-826(11)[40]		

6.5 The search for auto-ionizing states

Many auto-ionizing states have been observed from spectroscopic measurements of tin [171, 169, 170]. Using the 820 nm resonance laser step in the three-step ionization scheme presented in the previous section allowed the search for more AI states above the IP. Strong resonances were found within the region 12250 cm^{-1} to 12300 cm^{-1} ¹. Surprisingly, these resonances were present despite blocking the first two laser steps of the laser ionization scheme. Only a single laser achieved the resonant ionization of the tin beam at these frequencies. Using only the light from the grating Ti:Sa laser with 50 $\mu\text{J}/\text{pulse}$ a large scan region between 12040 cm^{-1} to 12888 cm^{-1} was investigated, four resonances were observed between 12250 cm^{-1} to 12300 cm^{-1} seen in Fig. 6.3, with the strongest resonance found at 12293.1 cm^{-1} . This resonance achieved about 10x greater ionization efficiency compared with the three-step ionization schemes presented in the previous section².

¹All provided wavenumbers are Doppler corrected to the rest frame of the atom.

²These measurements were performed in identical conditions as the search for AIs proceeded directly from the work presented in Sec. 6.4.

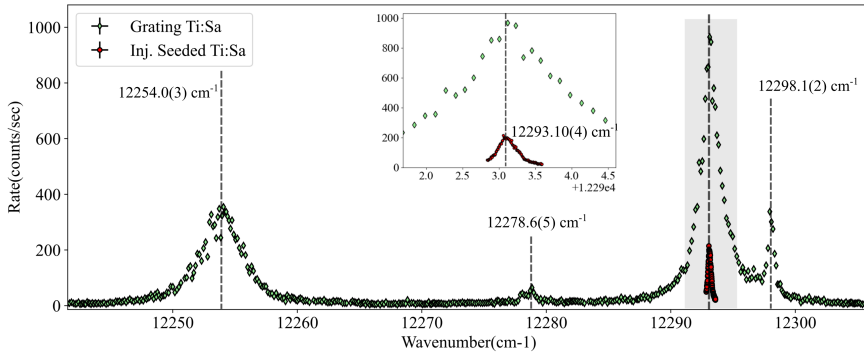


Figure 6.3: Observed resonances when using a single laser probing an atomic beam containing the stable tin isotopes, generated with the laser ablation ion source. The wavenumbers are Doppler corrected to the rest frame of the atom, using the mass of ^{120}Sn .

This resonance was studied further in high-resolution using the injection-seeded Ti:Sa laser, with as low as 60 nJ/pulse. Despite reducing the power a large FWHM of 7.6(3) GHz remained, which suggests a state with very short lifetime in the order of picoseconds, thus a transition to an AI. Assuming the AI state is between the IP at 59232.69 cm^{-1} to the highest lying measured AI at 63275.51 cm^{-1} the transitions would be expected from the populated states in the region between 46934 cm^{-1} to 50977 cm^{-1} or 34724 cm^{-1} to 38679 cm^{-1} assuming a 2-photon transition. Further investigation is needed into the origin of these efficient transitions as they could provide an efficient ionization scheme for future spectroscopic measurements. Despite the unclear origin, the simple laser ionization scheme was found useful during laser alignment and beam calibration due to its extraordinary ionization efficiency. These transitions were not further explored in this work as they exhibited no sensitivity to nuclear structure.

6.6 Ionization schemes considerations

In this work a large set of tin ionization schemes accessible using the CRIS technique have been investigated. Based on these measurements the criteria presented in the beginning of this chapter can be discussed. Neutralization calculations shown in Fig. 4.9 indicate a considerable population in the metastable states 1S_0 and 3P_2 using sodium neutralizer. The relative population in each state is difficult to measure accurately since the experiments were performed at different times under slightly different beam conditions. However,

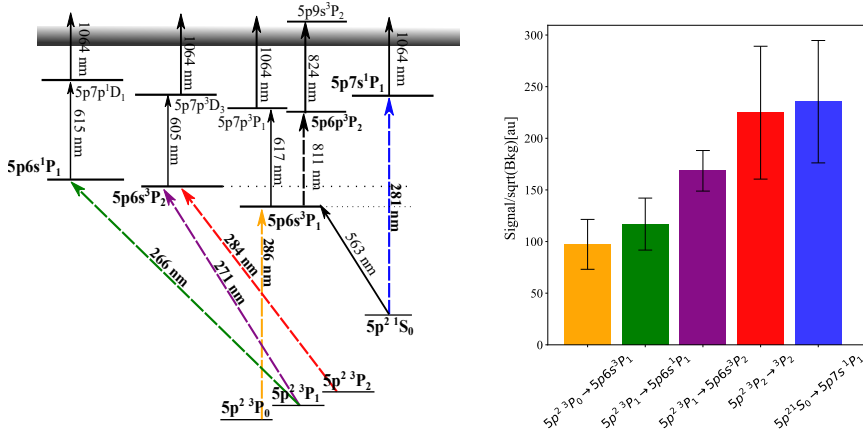
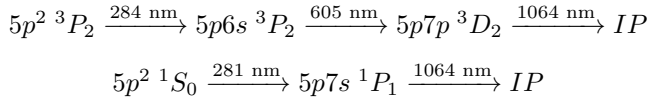


Figure 6.4: The studied ionization schemes shown to the left with the corresponding signal to background ratio for each investigated ionization scheme.

a series of measurements performed under similar conditions within the same experimental campaign (lasting 3 weeks) can be compared. In Fig. 6.4 the peak intensity of the ^{120}Sn resonance vs background rate is compared for the different ionization schemes. Under optimized conditions the greatest signal to background was achieved using the two resonance ionization schemes:



This confirms that the meta-stable states above the $5p^2\ ^3P_0$ ground-state carry a considerable population after the CEC when using sodium neutralizer, in agreement with the calculated atomic state population, seen in Fig. 4.9. Ionization schemes starting from the ground-state $5p^2\ ^3P_0$ resulted in a reduced signal to background by about half when compared to transitions from the $5p^2\ ^1S_0$ and $5p^2\ ^3P_2$ atomic states. As discussed in Sec. 6.4, the multi-step ionization scheme to the observed AI did not provide any observed enhancement in efficiency. A large background signal was observed when using 800 nm light, which could be caused by both resonant or non-resonant ionization from high-lying populated states, as revealed from the efficient ionization achieved using a single laser step as presented in Sec. 6.5.

The saturation curves for the resonant steps in the transitions from 1S_0 and 3P_2 reveal the resonance count rate with increased laser laser fluence, see Fig. 6.5. Additional laser power beyond the saturation level of the transition result

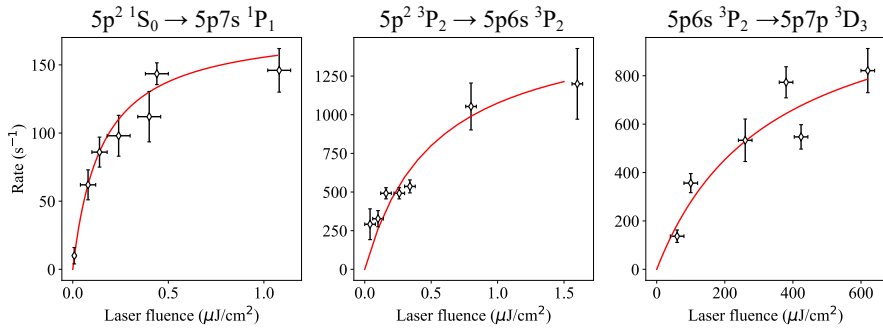


Figure 6.5: Measured saturation curves of atomic transitions in tin.

in power broadening of the resonance linewidth, reducing the quality of the hyperfine structure measurement.

The sensitivity to nuclear structure properties of the studied atomic levels and transitions have been determined, as presented Ref. [107]. While all probed transitions from the $^3P_{0,1,2}$ states have a similar sensitivity to the nuclear charge radius, a slightly reduced sensitivity was observed for the $^1S_0 \xrightarrow{281\text{ nm}} ^1P_1$ transition. Considering the sensitivity to the nuclear moments, the $5p6s\ ^1P_1$ state remains the most sensitive to the magnetic dipole moment. However, the $5p6s\ ^3P_2$ state is most sensitive to the electric quadrupole moment while maintaining a comparable sensitivity to the magnetic dipole moment. Additionally, the greater electronic angular momentum number for the $J = 2 \rightarrow J' = 2$ transition is favorable for nuclear spin determination³. Consequently, the sensitivity to nuclear structure together with the large relative atomic population in the $5p^2\ ^3P_2$ state makes the $5p^2\ ^3P_2 \xrightarrow{284\text{ nm}} 5p6s\ ^3P_2$ transition favorable for probing the nuclear structure of the neutron-deficient tin isotopes.

³As described in Sec. 7.9.

Chapter 7

Measurements of the short-lived tin isotopes

This chapter presents the results from the experimental campaign dedicated to the production and laser spectroscopy of neutron-deficient tin isotopes using the CRIS technique at ISOLDE. Two newly developed ionization schemes discussed in the previous chapter were used for probing the hyperfine structures, namely $5p^2\ ^3P_2 \xrightarrow{284\text{nm}} 5p6s\ ^3P_2(\text{PP})$ and $5p^2\ ^1S_0 \xrightarrow{281\text{nm}} 5p7s\ ^1P_1(\text{SP})$. The hyperfine structures were successfully resolved for 21 tin isotopes ranging from ^{124}Sn down to ^{104}Sn , providing the first laser spectroscopic measurements of $^{104,105,106,107}\text{Sn}$. The analysis of the acquired data is presented and the nuclear observables (spin, moments and charge radii) of interest in this work are revealed.

7.1 Experimental conditions and resolved spectra

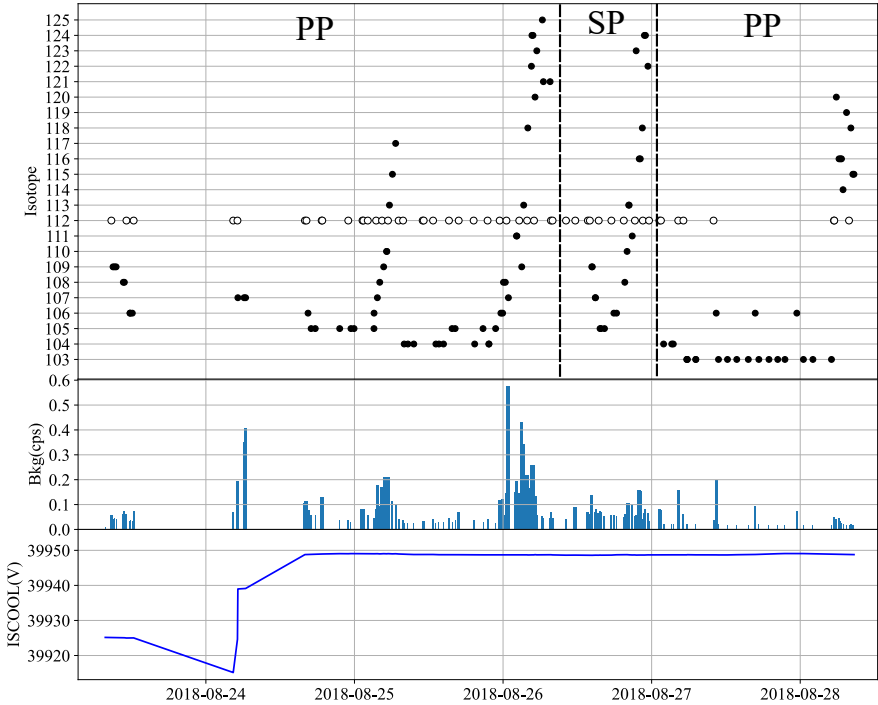


Figure 7.1: The sequence of measured tin isotopes over the duration of the experiment are presented in the top figure. ^{112}Sn was selected as a reference isotope and was therefore measured repeatedly indicated with white dots. Below is the ionizing radiation related background rate registered at the MagneTOF ion detector (see text for details) and below that is the measured ISCOOL platform voltage during the course of the experiment.

The experimental campaign at ISOLDE dedicated to the production and laser spectroscopy of neutron-deficient tin isotopes was performed in August 2018 from 23/08/2018 to 28/08/2018. Each selected isotope was measured independently in sequence over the duration of the experiment as indicated in Fig. 7.1. The lightest stable tin isotope ^{112}Sn was selected as reference for isotope shift determinations, and measured repeatedly throughout the campaign to correct for drifts and monitor any changes in the line-profile occurring during the experiment. The radiation related background associated with the

radioactive beam content sent to CRIS is presented in the figure¹. Initially, significant transmission losses were observed across ISCOOL with less than <10% transmission measured using Faraday cups before and after the cooler. In order to improve the efficiency the ISCOOL electrodes were fine tuned and the platform voltage was changed from 39925.2 to 39948.8V, where it remained stable within $\sigma = 0.3$ V over the course of the experiment, see Fig. 7.1. After this intervention the transmission efficiency of neutron-deficient tin isotopes was enhanced from 20 and up to 40% for the lighter masses. Only data acquired with the stable ISCOOL settings have been used to extract nuclear structure information. The tin ions sent into the CRIS beamline from ISCOOL were neutralized in the CEC containing potassium vapour maintained at a temperature of 200(20)°C. The PP transition was optimized on resonance with a laser fluence of 100(40) nJ/cm² with the second step in the PP transition scheme at 600(80) μ J/cm². The SP transition was probed using the two step scheme with a laser fluence of 300(50) nJ/cm² for the resonant step, see saturation curves presented in Fig. 6.5. Due to reduced ionization efficiency when using the SP transition scheme (Fig. 7.3) the lasers were switched back for probing the PP transition for the remainder of the experiment. The reduced efficiency of the SP transition was partly attributed to the use of potassium neutralizer where a lower population is expected in the 1S_0 state according to simulations, see Fig. 4.10².

¹This was determined by gating on events registered in the ion detector 5-10 ms after the ion bunch arrival.

²These newer simulations were not available at the time of the experiment.

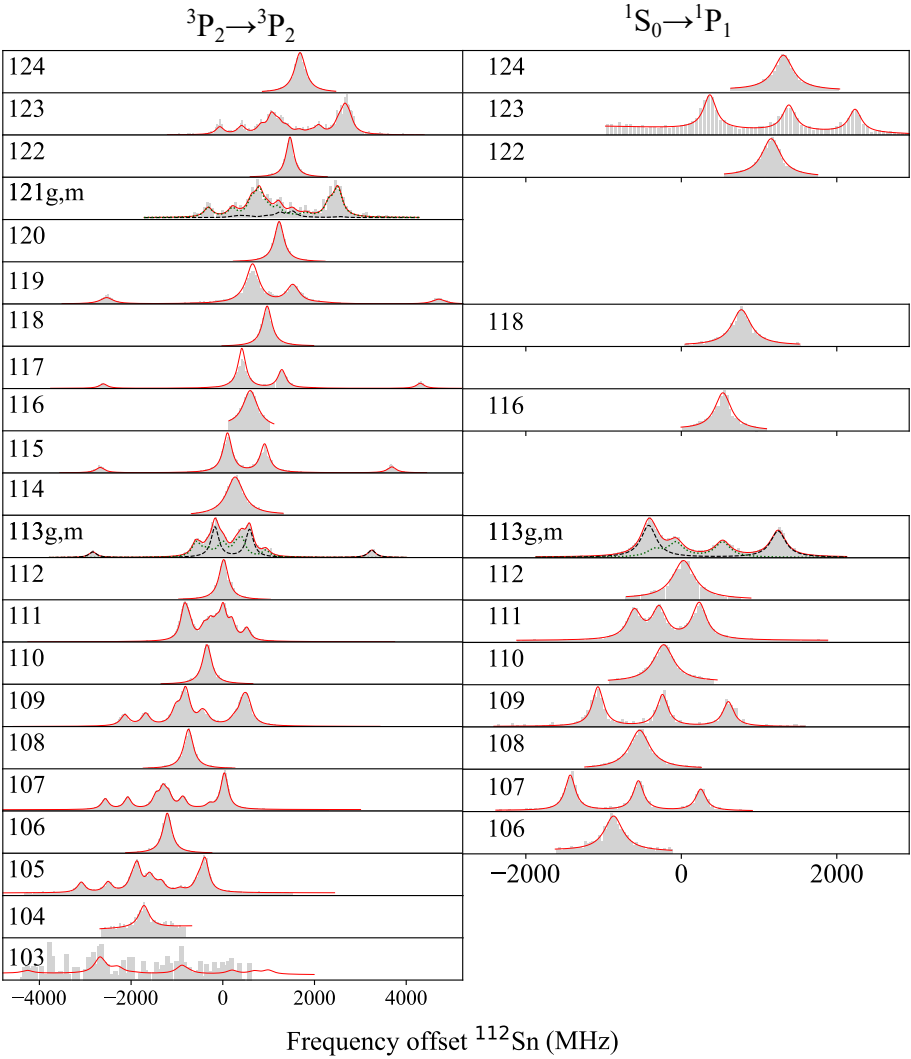


Figure 7.2: Overview of the tin spectra probed using the PP and SP transition schemes. The fitted functions are indicated with a red line. Isomers and ground states are displayed with dotted green and dashed black lines respectively. The spectrum of ^{103}Sn is discussed later in Sec. 9.1.

The studied tin isotopes probed using the PP and SP transitions are presented in Fig. 7.2. While the sequence $^{124-104}\text{Sn}$ were fully measured using the PP transition scheme, a few isotopes were missed when scanning the SP transition due to time constraints. All spectra have been Doppler corrected with respect to their mass provided in Tab. 7.1, therefore the relative shift in centroid position with respect to the ^{112}Sn is a clear indication of the isotope shift. The spin 0 isotopes can be easily distinguished from the lacking hyperfine structure splitting, the odd-even tin isotopes show multiple peaks containing the information of their nuclear moment and spin properties. Some isotopes have known long-lived isomers, as indicated in Tab. 7.1. The structure observed when scanning ^{121}Sn was dominated by its long-lived spin 11/2 isomeric state with the ground-state convoluted within the spectrum. While the spin 3/2 isomer in ^{123}Sn and the spin 7/2 isomer in ^{117}Sn could not clearly be distinguished, a small bump was observed in the spectrum of ^{119}Sn which was likely originating from the presence of the spin 11/2 isomeric state. A spin 7/2 isomer expected in ^{113}Sn was seen convoluted with the spin 1/2 ground state in both transitions. The isomer corresponded to roughly 30% of the ground state yield, the nuclear moments and radii of this isomer were unknown prior to this work. A significant time of the experiment was dedicated to search for ^{103}Sn resonances using the PP transition scheme. In order to maximize efficiency for this endeavor the laser fluence of the first step was increased to 300(50) nJ/cm² which increased the linewidth from 200(20) to 450(40) MHz but enhanced the efficiency with roughly 30%. While hints of a ^{103}Sn were likely observed, the hyperfine structure could not be fully resolved due to the large amount of isobaric contamination and low yield, discussed further in Chp. 9.

The peak rate of the largest resonance peak, background rate and their signal to background ratio $S/\sqrt{BG}$ are displayed in the bar charts in Fig. 7.3 for all studied spectra in both transitions. A higher overall signal to background was observed using the PP transition scheme. The efficiency from ISCOOL to resonant detection at CRIS, discussed in Sec. 4.9, was estimated over the course of the experiment. The ion current of a mass separated beam of stable ^{112}Sn was measured after release from ISCOOL. The Faraday cup was then removed and the beam was sent to CRIS where the peak rate of resonantly ionized ions were measured at the end of the beamline. The CRIS efficiency was determined from the ratio of the peak rate at CRIS compared over the ion rate measured at the ISCOOL Faraday cup. The efficiency of 1/1200(500) was achieved on average during the experiment using the PP transition. A reduced efficiency of 1/7000 was measured with a single measurement using the SP transition due to the lower laser ionization efficiency.

There was a clear reduction in yield towards more neutron-deficient isotopes with measured rates of only 4 cps on resonance for ^{104}Sn with a background of

1 cps. The suggested ^{103}Sn resonance peak had a signal rate of 0.1 cps on top of a background rate of 0.4 cps which assuming the average CRIS efficiency of $1/1200(500)$ results in $120(50)$ ions/s released after ISCOOL. The estimated yields from these measurements are provided in Tab. 7.1, which are in good agreement with the yields estimated in Tab. 4.1.

While the signal rate drops exponentially with reducing neutron number the background rate follows a slower decline which results in a rapidly reduced signal to background for the most exotic cases. The beam current for every mass was measured after ISCOOL using a Faraday cup. By measuring the beam current with RILIS both on and off, the non-resonantly ionized isobaric beam contamination passing the HRS magnet could be identified. The contamination % in the beam at different masses are displayed in grey in Fig. 7.3 indicating that an increasing majority of the beam content was composed of isobaric contamination for the neutron-deficient tin isotopes. A more detailed discussion of the origin of this background is provided in Chp. 9.

Table 7.1: Overview of tin isotopes and isomers relevant in this work, the provided masses and half-lives are taken from [176]. Ion yields after ISCOOL have been estimated from peak count rates on resonance measured at CRIS (using the PP transition scheme) assuming an efficiency of 1/1200(500). The number of measurements of each mass using the SP and PP transition scheme are provided.

Isotope	Half-Life	I^π	M(amu)	Yield(ions/s)	# runs(PP)	# runs(SP)
103	1.7 s	$(\frac{5}{2}^+)$	102.928105(76)	120(50)	16	0
104	3.8 s	0	103.9231052(6)	$30(12) \cdot 10^2$	12	0
105	7 s	$(\frac{5}{2}^+)$	104.9212684(4)	$52(22) \cdot 10^3$	10	3
106	20.8 s	0	105.9169574(5)	$26(10) \cdot 10^4$	10	2
107	32.7 s	$(\frac{5}{2}^+)$	106.9157137(6)	$34(14) \cdot 10^5$	5	2
108	115 s	0	107.9118943(6)	$40(17) \cdot 10^6$	5	1
109	2.9 m	$\frac{5}{2}^+$	108.9112928(9)	$1.4(6) \cdot 10^8$	5	2
110	10.3 m	0	109.9078448(15)	$> 10^8$	2	1
111	18.1 m	$\frac{7}{2}^+$	110.9077411(6)		2	1
112	STABLE	0	111.90482387(1)		40	10
113	115 d	$\frac{1}{2}^+$	112.9051758(2)		2	2
113m	24 m	$\frac{7}{2}^+$	-			
114	STABLE	0	113.9027801(3)		1	
115	STABLE	$\frac{1}{2}^+$	114.9033447(0)		3	
115m	3.26 μ s	$\frac{7}{2}^+$				
116	STABLE	0	115.9017428(0)		3	2
117	STABLE	$\frac{1}{2}^+$	116.902954(1)		1	
117m	14 d	$\frac{11}{2}^-$				
118	STABLE	0	117.9016066(1)		2	1
119	STABLE	$\frac{1}{2}^+$	118.9033112(1)		1	
119m	293 d	$\frac{11}{2}^-$	-			
120	STABLE	0	119.9022019(1)		2	
121	27 d	$\frac{3}{2}^+$	120.9042428(1)		2	
121m	43.9 y	$\frac{11}{2}^-$	-			
122	STABLE	0	121.903444(3)		1	1
123	129 d	$\frac{11}{2}^-$	122.9057254(3)		1	1
123m	40 m	$\frac{3}{2}^+$	-			
124	STABLE	0	123.9052767(1)		2	2

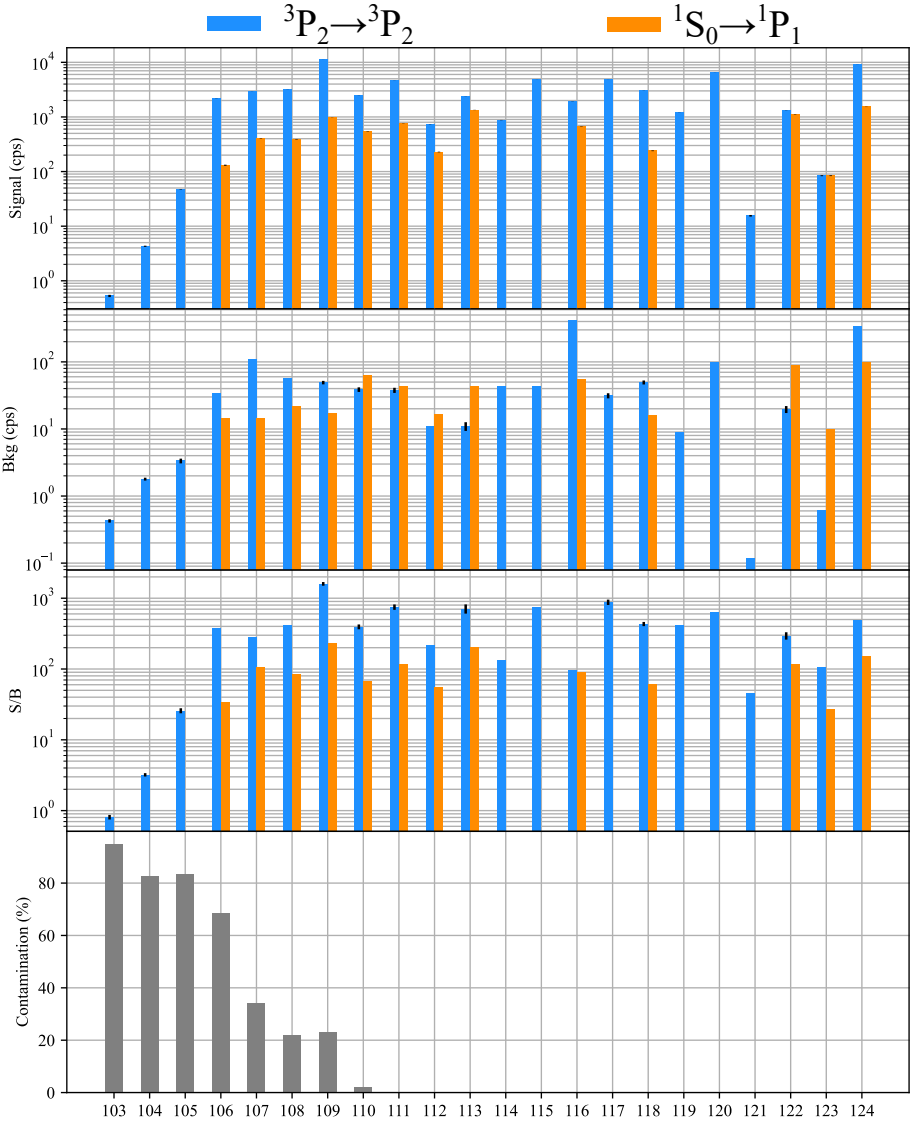


Figure 7.3: Overview of the signal rate and background for each tin isotope measured at CRIS. The bottom bar chart shows the percentage of non-resonantly ionized isobars in the Sn beam of a given mass, entering the ISCOOL buncher (see details in the text).

7.2 Characterisation of the reference isotope, ^{112}Sn

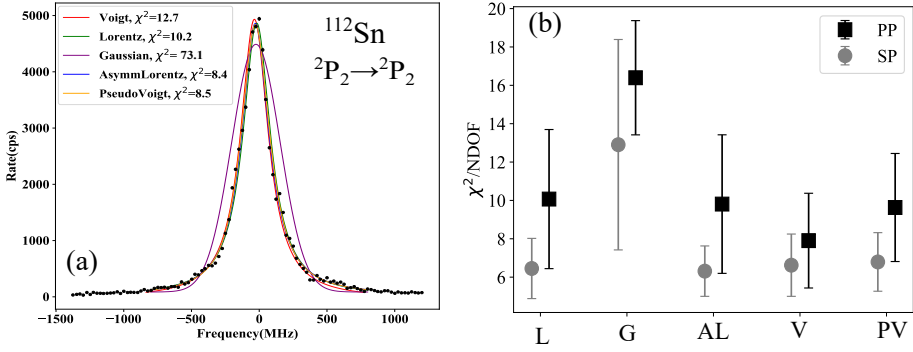


Figure 7.4: Figure (a) show a sample spectrum of ^{112}Sn fitted using different peak profiles. Figure (b) show the average reduced χ^2 of the fitted reference resonances, using the different peak profiles: Lorentz (L), Gaussian (G), Voigt (V) PseudoVoigt (PV) and Asymmetric Lorentz (AL).

A sample spectrum of the spin 0 reference isotope ^{112}Sn fitted using different peak profiles, Lorentz (L), Gaussian (G), Voigt (V) PseudoVoigt (PV) and Asymmetric Lorentz (AL), is displayed in Fig. 7.4(a). The average goodness-of-fit, defined by $\chi^2/NDOF$, was determined from fitting all reference spectra, and is presented for the different peak profiles in Fig. 7.4(b). While the Gaussian profile could be eliminated directly from observation (see fit in Fig. 7.4), there was no observable change in $\chi^2/NDOF$ between the other fit functions. There was no clear indication of peak asymmetry or side-peak distortions in the spectra, while both PV and the AL profiles contain the additional asymmetric degree of freedom, there was no observable change in $\chi^2/NDOF$ between the other fit functions. Notably, the Gaussian contribution was negligible using the Voigt profile, hence the Lorentz profile is favored.

The centroid frequency of ^{112}Sn was studied over the course of the experiment to investigate drift in the measured reference frequency. The measured frequency drifted roughly 100 MHz in 90h during the experiment, which was well correlated with the measured reference diode frequency, see Fig. 7.5. This drift was corrected for by minimizing the spread with respect to an α -correction parameter, as described in Sec. 5.2. The standard deviation of the measured ^{112}Sn centroid frequencies after this correction procedure are seen in Fig. 7.5(a). This procedure was performed on the extracted centroid frequencies assuming the different peak profiles, which indicated that the scatter was minimized when fitting using a Lorentz profile with an α -correction parameter of 1.49. The

measured spread in reference frequency was reduced to only 8.5 MHz from the 100 MHz drift observed before correction. This observation of minimized spread after correction together with the indifference to the goodness-of-fit between the different peak profiles motivated the use of a Lorentz profile for fitting the hyperfine spectra using the PP transition. This drift correction using $\alpha = 1.49$ was performed on all spectra acquired using the PP transition scheme.

This drift correction was not needed for the resonances probing the SP transition since no measurable drift beyond the measured ^{112}Sn reference centroid error could be observed, due to the short time duration of these measurements. Likewise, a Lorentz profile was used for the fitting of these measurements since the other peak profiles provided no improvement in the goodness-of-fit.

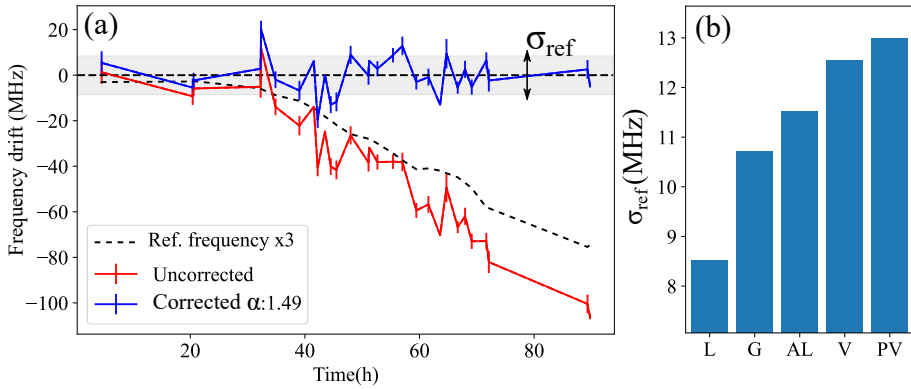


Figure 7.5: The measured centroid frequency of ^{112}Sn drifting over the course of the experiment in correlation with the measured reference diode frequency drift is shown in (a) together with the frequency after the correction procedure. The standard deviation of the spread after correction are displayed in (b) for the different assumed fit profiles.

The TOF information acquired with each measurements is demonstrated with a time-sliced sample spectrum in Fig. 7.6(a) together with the corresponding TOF histogram in 7.6(b). This TOF histogram provides an interesting snapshot of the distribution of ions within the CRIS beamline when the resonance ionization process occurs. The TOF histogram shows a smooth ion distribution with a clear cutoff at 8 μs . This cut is a result of the deflection of ions occurring at the electrostatic deflector plate before the interaction region. This indicates that a part of the neutralized tin beam was still located before the interaction region when the resonance ionization process occurred, such that these (slower) resonantly ionized ions are deflected before reaching the interaction region.

This highlight the possible gain in efficiency that can be achieved with better beam optics at CRIS. A small amount of ions arrived at a greater TOF which indicate that not all ions were removed before the interaction region by the deflector plate. These ions could contribute to the detected background signal. Furthermore, a spike is observed shortly before the deflector plate cutoff, this could be caused by beam steering of the resonantly ionized ions by the stray field from the deflector plate. During these measurements the timing of the ion bunch release from ISCOOL was optimized with respect to maximized efficiency at CRIS. The peak efficiency is often observed when the laser beam overlap takes place closer to the deflector plate in the interaction region. This is due to the diverging ion beam along the CRIS beamline due to lack of focusing elements before the CEC. These observations highlight the potential gain that could be achieved with improved beam optics at the CRIS beamline.

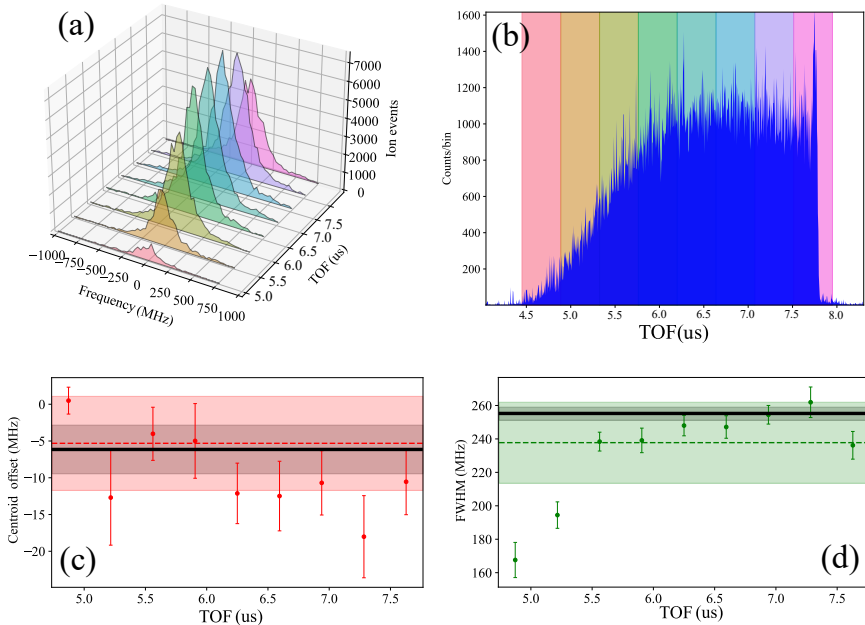


Figure 7.6: (a) Sample spectrum of ^{112}Sn sliced across the TOF with 500 ns TOF slices indicated in the TOF histogram in (b). (c) show the variation of centroid frequency with respect to TOF, indicated in red with red dashed line showing the weighted mean. (d) show the change in FWHM with respect to TOF. The centroid position and FWHM when gating on the complete TOF are indicated with black dashed lines together with grey shaded uncertainty.

The frequency spectrum was gated with 500 ns timeslices as seen in figure 7.6(a) and each slice was fitted for studying changes occurring with different TOF. The dependence of centroid frequency and FWHM with respect to TOF was investigated and compared with the centroid frequency and FWHM of the resonance when gating on the full TOF bunch, seen in Fig. 7.6(c) and (d). While there was no clear indication of a change in centroid frequency with respect to different TOF slices, a trend of reduced FWHM was observed when gating on the shorter TOF. This was likely caused by the reduced power broadening resulting from a lower laser fluence further down the beampath. This indicated that there was considerable power broadening of the spectra during the experiment where an average linewidth of 250(30)MHz was measured during the initial phase of the experiment when probing the PP transition. The reduced statistics from gating on a narrow TOF region was not considered worth the benefit of potential improved resolution, a 4 μs TOF gate was therefore used when fitting all spectra.

7.3 Hyperfine structure considerations

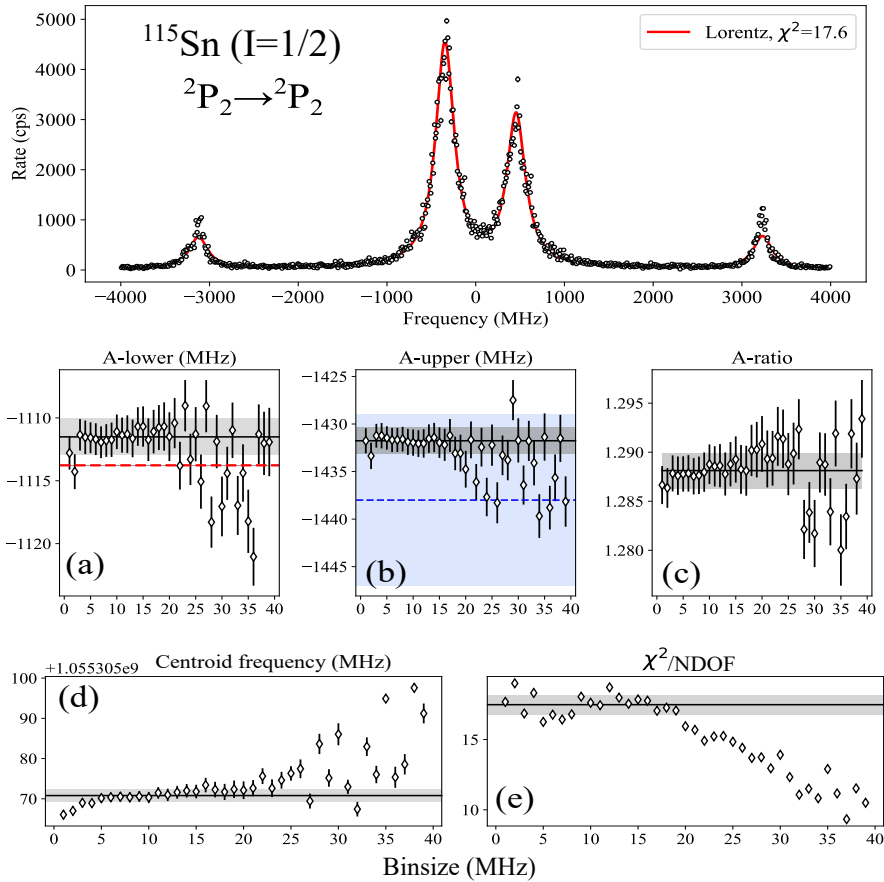


Figure 7.7: Fitted hyperfine spectrum of ^{115}Sn at varying bin size from 1 to 40 MHz. The different parameters extracted from the fit are displayed in the subfigures a-e showing the convergence to the final extracted parameter values with respect to bin size indicated with solid black lines with grey shaded errors. The A_{hf} -values from Ref. [173, 107] are displayed with dashed lines in (a) and (b), the literature error in (a) is smaller than the line.

The measured spectra of the odd-even tin isotopes exhibit hyperfine structures which contain properties of their spin and nuclear moments. These structures were fitted using Lorentzian profiles with shared FWHM for every resonance peak observed in the spectrum, including any present resonances from isomer

states. The hyperfine structures were fitted using different bin sizes in order to determine the influence of the selected bin size on the extracted values. A sample spectrum of the spin 1/2 isotopes ^{115}Sn scanned at 5 MHz/s and presented with 15 MHz bin size is fitted in Fig. 7.7 together with the corresponding fit parameter values for different bin sizes ranging from 1 to 40 MHz in 1 MHz increments. A large systematic scatter in the extracted values was observed when fitting with bin sizes greater than 20 MHz, which converged below 20 MHz and started to deviate again below 5 MHz as seen in Fig. 7.7. The average value is selected from fit parameters extracted using bin sizes between 10 and 20 MHz indicated with dashed line with shaded errors. The increased $\chi^2/NDOF$ when reducing the bin size reveal that the fluctuations in beam/laser intensity are not averaged properly within the smaller bins. The errors of the fitted parameters are therefore scaled with $\chi^2/NDOF$ to account for this systematic uncertainty, as described in Sec. 5.

The hyperfine structures observed using the PP transition are particularly complex for $I > 1/2$ isotopes, where many resonance peaks are overlapped within the observed spectra, see Fig. 7.2. For example the structure of ^{111}Sn with spin 7/2 contains 13 resonance peaks. In order to fit these complex structures the ratio of the hyperfine A_{hf} and B_{hf} parameters needed to be constrained.

7.3.1 A_{hf} -ratios

The three stable spin 1/2 isotopes $^{119,117,115}\text{Sn}$ provide clean references for the hyperfine A_{hf} -values without influence from the electric quadrupole moment. The extracted A_{hf} -values of the upper (u) and lower (l) levels from fitting these structures were in good agreement with literature and the offline work, seen in Tab. 7.2. The weighted-mean A_{hf} -ratio of the upper and lower atomic levels for the PP transition $A_{hf}^{(u)}/A_{hf}^{(l)}$ was determined, as presented in Tab. 7.2. The determined ratio was in excellent agreement with the calculated A_{hf} -ratio using the FSCC approach and those experimentally determined from the offline measurements, see Fig. 7.8(a). The final A-ratio of 1.2817(20) was used to constrain the fitting of the spectrum probed using the PP transition. The hyperfine anomaly is neglected in this analysis as it is expected to be smaller than the experimental uncertainty. The hyperfine anomaly for the $5p^2\ ^3P_2$ state for ^{115}Sn and ^{117}Sn account to a difference of 0.0003(10)% in the A_{hf} values [177]. Furthermore, in the work by D.T. Yordanov *et al.* [77] the hyperfine anomaly was expected to account for a difference of less than 0.05% between ^{119}Sn and ^{131}Sn . A further investigation into the influence of the hyperfine anomaly of the probed states in this work might be considered but are not expected to influence the results in this work.

Table 7.2: A_{hf} -values for the upper(u) and lower(l) level of the PP transition for the spin 1/2 tin isotopes.

Isotope	$A_{hf}^{(l)}$ [MHz]	$A_{ref}^{(l)}$ [MHz] [173]	$A_{hf}^{(u)}$ [MHz]	$A_{ref}^{(u)}$ [MHz] [107]	A_{ratio}
115	-1112(2)	-1113.770(4)	-1432(1)	-1438(9)	1.2871(39)
117	-1213(3)	-1212.956(3)	-1554(3)	-1546(9)	1.2805(34)
119	-1272(3)	-1269.419(3)	-1627(3)	-1621(6)	1.2790(35)
Online:					1.2819(21)
Offline:					1.2790(89)
Final:					1.2817(20)
FSCC[107]:					1.309(56)

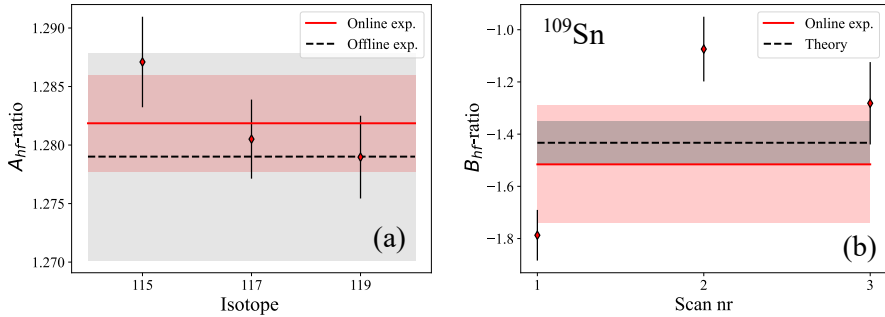


Figure 7.8: (a) shows the A_{hf} -ratios from measurements of spin 1/2 isotopes together with the weighted average indicated in red, the A_{hf} -ratio from the offline measurements is shown with black dashed line with errors indicated with grey shade. (b) shows the B_{hf} -ratios from three separate measurements of ^{109}Sn ($I=5/2$) together with the weighted average indicated with a red line with errors indicated with red shade. The calculated B_{hf} -ratio with uncertainty is indicated with a black dashed line with grey shaded error.

7.3.2 B_{hf} -ratios

The calculated EFG of two levels involved in the PP transition provide a theoretical estimate of the B_{hf} -ratio of -1.43(8)[107]. The hyperfine structures probed with the PP transition allow testing the accuracy of these theoretical calculations. One particularly suitable candidate for testing the B-ratio was ^{109}Sn with spin 5/2, which has been measured using different laser spectroscopy techniques [174, 77]. The hyperfine structure of ^{109}Sn was measured in three

separate measurements during the experiment. These spectra were fitted with constrained A_{hf} -ratio determined in the previous section, the resulting B_{hf} -values were used for calculating the B_{hf} -ratio, as presented in 7.3 and shown in Fig. 7.8(b). The considerable scatter of the extracted B_{hf} -ratios from the different measurements highlighted the need for additional constraints. The weighted-mean B_{hf} -ratio with scaled errors from these measurements, however, was in good agreement with the calculated B-ratio value using the FSCC approach, with an estimated uncertainty significantly smaller than the experimentally determined. The theoretically calculated B_{hf} -ratio with its estimated uncertainty was used to constrain the fit of the hyperfine structures measured using the PP transition.

Table 7.3: The table show the measured B_{hf} -values for the upper and lower atomic levels involved in the PP transition probing ^{109}Sn . The B_{hf} -ratios are presented for each measurement including the weighted mean with scaled errors. The Experimental error is compared with that calculated using FSCC from ref. [107].

Scan	$B_{hf}^{(l)}$ [MHz]	$B_{hf}^{(u)}$ [MHz]	B_{hf} -ratio
1	-141(10)	252(9)	-1.79(8)
2	-210(22)	225(14)	-1.07(12)
3	-192(26)	245(15)	-1.28(15)
B_{hf} -ratio(Exp.):			-1.52(23)
B_{hf} -ratio(Theory)[107]:			-1.43(8)

7.4 Spin assignment of $^{105,107}\text{Sn}$

The observed hyperfine structures of $^{105,107}\text{Sn}$ contain information of their nuclear spin, which prior to this work remained only tentatively assigned as $I^\pi = (5/2^+)$ from gamma spectroscopy of subsequent alpha-decays from neutron-deficient Tellurium isotopes [22] and single neutron knockout studies [178]. The hyperfine structures of these isotopes contained multiple overlapped peaks in the PP transition. This clearly excludes the four peak structure expected assuming spin 1/2. In order to identify higher spins a more sophisticated treatment is required. Therefore, the hyperfine structures were fitted with model functions with different assumed nuclear spin I , with constrained A_{hf} and B_{hf} -ratios defined in earlier sections, as well as fixed peak intensity ratios according to the Racah intensities. The goodness-of-fit $\chi^2/NDOF$ was investigated after fitting each model to the hyperfine structure to determine the spin which best fit the observed structure. The uncertainty in $\chi^2/NDOF$ was determined from

the standard deviation of the scatter of $\chi^2/NDOF$ by fitting the structure at different bin sizes from 15 to 20 MHz in 0.5 MHz increments as done for ^{115}Sn in Fig. 7.7(e). Since the nuclear spin of ^{111}Sn and ^{109}Sn have been firmly assigned as 7/2 and 5/2 respectively, they could be used to test the reliability of this approach, see Fig. 7.9. This procedure could correctly reveal a minimum in $\chi^2/NDOF$ at $I = 7/2$ for ^{111}Sn and $I = 5/2$ for ^{109}Sn . However, while $I = 5/2$ could be excluded from ^{111}Sn , $I = 9/2$ was within 2σ of the minimum value and could not be excluded using this method. The amplitude of the central peak in the hyperfine structure of ^{111}Sn could not be reproduced with the fixed Racah intensities, despite changing spin of the fitted model. The strongly convoluted hyperfine structure means it is particularly sensitive to any uncertainty in the fixed hyperfine ratios, which is a likely reason for this offset. However, the nuclear spin 5/2 for ^{109}Sn could be determined with good accuracy which provides confidence in the approach for distinguishing spin 5/2 structures. This approach was applied to the observed structures of $^{107,105}\text{Sn}$ which indicated spin 5/2 for both isotopes with above 5σ confidence.

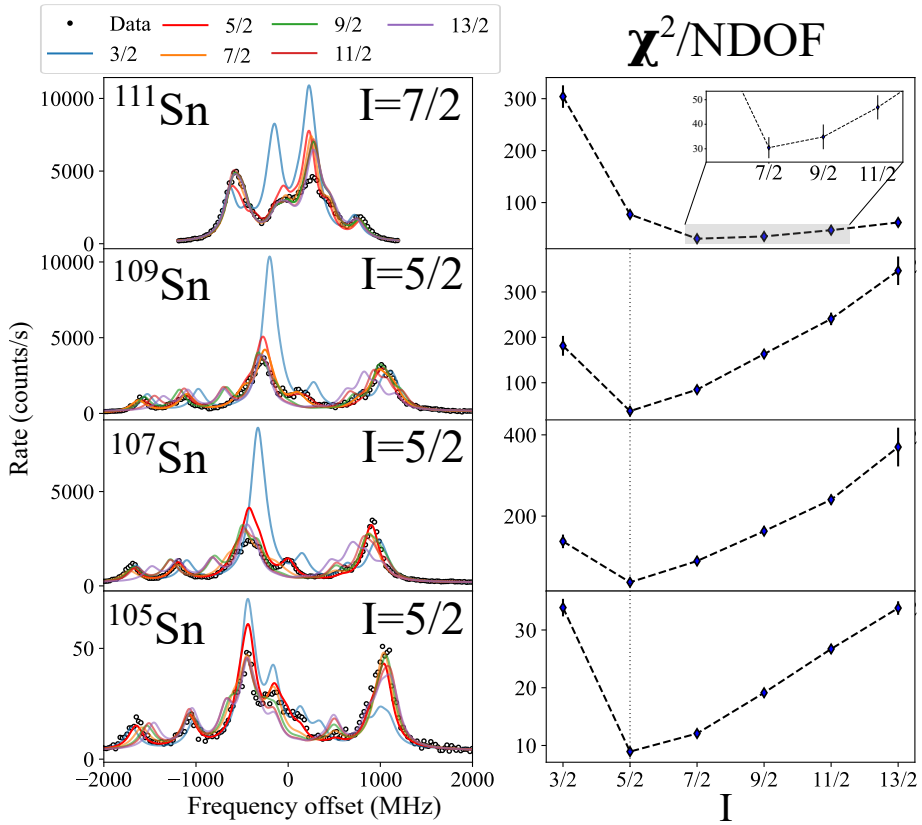


Figure 7.9: The figure shows the spin assignment procedure used to determine the spin of ^{107}Sn and ^{105}Sn . With fixed Racah intensities, A_{hf} and B_{hf} -ratios the goodness-of-fit is determined at different assumed nuclear spin I indicating a minimum at spin 5/2 for these isotopes.

7.5 Hyperfine parameters and nuclear moments

The hyperfine structures of the odd-even isotopes fitted with fixed A_{hf} , B_{hf} -ratios are displayed in Fig. 7.2. While no isomers were observed below ^{112}Sn , a spin 7/2 isomer known to exist in ^{113}Sn , was seen overlapped with the spin 1/2 ground-state in both transitions. Since the nuclear moments of the ground-state are known from literature (Ref. [85]) the spin 1/2 hyperfine structure could be constrained, in order to extract the hyperfine structure of the spin 7/2 isomeric state. Likewise, the fitted hyperfine structure for ^{121}Sn was constrained using

recalculated with hyperfine fields provided in Paper I together with nuclear moments provided from Ref. [77]. The A_{hf} and B_{hf} values extracted from the fitted spectra are provided in Tab. 7.4 and 7.5 respectively. The recalculated values are presented in italics.

The nuclear magnetic moments were calculated from the hyperfine A_{hf} -values, knowing the nuclear spin together with the hyperfine magnetic fields of the lower state of the PP transition $A_0^{(l)}(PP) = 607.50(4)$ MHz and the upper state for the SP transition $A_0^{(u)}(SP) = 638(8)$ MHz, as presented in paper I. The weighted-mean value of the magnetic moments extracted from the two measured A_{hf} -values, with errors not reduced below minimal sample error, are presented in Tab. 7.4 together with literature values from other laser spectroscopy measurements by D.T Yordanov *et al.*, J. Eberz *et al.* [77, 76] and from Ivanov *et al.* [179] indicated in black. The g-factors can be grouped according to the nuclear spin of the state. This illustrates the sensitivity of the g-factor to the unpaired valence neutron, which is different for the spin 5/2, 7/2 and 1/2 states, as discussed later in 8.1. The experimental g-factors $g = \mu_I/I$ are shown in Fig. 7.10 which indicate good agreement within 2σ of the literature values.

Table 7.4: A_{hf} -values (in MHz) from the measured hyperfine structures using the SP and PP transitions. The weighted-mean magnetic moment was calculated using hyperfine magnetic fields from paper I with the A_{hf} -values from both transitions. The A_{hf} -values shown in italics have been calculated from literature moments (see text for details).

Isotope	I	PP		SP	$\mu^{Exp.}(\mu_N)$	$\mu^{lit.}[\mu_N]$ [Ref.]
		$A_{hf}^{(l)}$	$A_{hf}^{(u)}$	$A_{hf}^{(u)}$		
105	5/2 ⁺	-264(1)	-339(2)		-1.088(6)	
107	5/2 ⁺	-262(1)	-336(1)	-272(2)	-1.073(6)	
109	5/2 ⁺	-264(2)	-338(2)	-274(2)	-1.081(7)	-1.081(1) [77]
111	7/2 ⁺	104(2)	133(2)	106(2)	0.590(10)	0.608(4) [76]
113	1/2 ⁺	<i>-1066(1)</i>	<i>-1368(2)</i>	<i>-1119(14)</i>		-0.8771(1) [85]
113m	7/2 ⁺	114(3)	146(3)	117(5)	0.651(13)	
115	1/2 ⁺	-1111(2)	-1433(3)		-0.915(2)	-0.9167(1) [77]
115m	7/2 ⁺					0.683(10) [179]
117	1/2 ⁺	-1214(3)	-1553(3)		-0.999(3)	-0.9983(1) [77]
119	1/2 ⁺	-1271(3)	-1626(3)		-1.046(3)	-1.0448(1) [77]
121	3/2 ⁺	<i>283(0)</i>	<i>362(4)</i>			0.698(1) [77]
121m	11/2 ⁻	<i>-154(0)</i>	<i>-196(2)</i>			-1.394(1) [77]
123	11/2 ⁻	-151(2)	-194(3)	-157(2)	-1.363(18)	-1.347(1) [77]

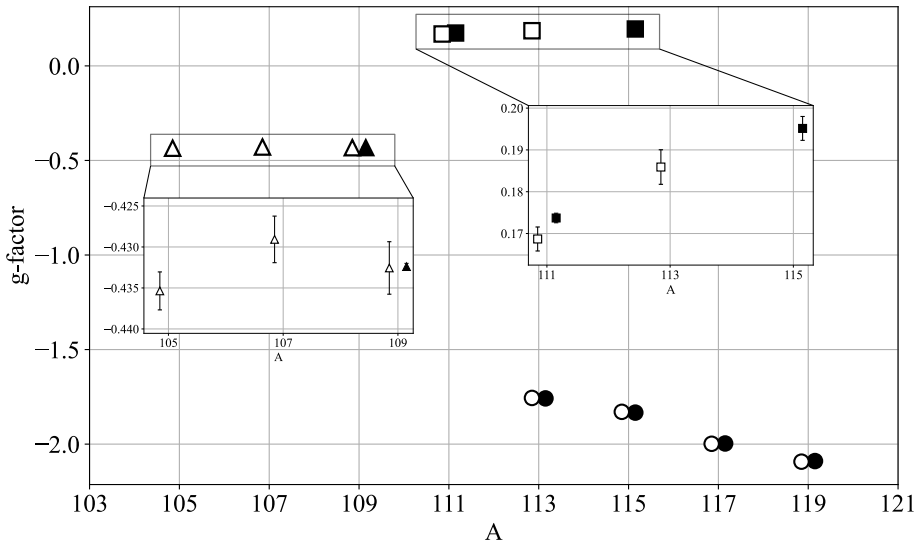


Figure 7.10: Experimental g-factors of the $5/2^+$, $7/2^+$ and $1/2^+$ states of tin isotopes indicated with triangles, squares and circles respectively. The data from this work is indicated in white with literature values from Ref. [77, 85, 179, 76] in black.

The spectroscopic quadrupole moments were extracted from the experimental B_{hf} values using the EFG for the upper state in the PP transition $q_0^{(u)}(PP) = 990(40)$ MHz/b and the upper state for the SP transition $A_0^{(u)}(SP) = 378(68)$ MHz/b, as presented in paper I³. The weighted-mean Q_s from the probed atomic levels where $J > 1/2$, with errors not reduced below minimal sample error, are presented in Tab. 7.5. The statistical and systematic errors (resulting from the EFG) are separately shown in the table. The Q_s values with their total uncertainties are compared with literature in Fig. 7.11. The minor discrepancy of Q_s in ^{109}Sn between this work and that from [77] could be explained by the different calculated EFGs.

The spin $5/2^+$ isotopes reveal a linearly decreasing quadrupole moment with reduced neutron number down to ^{105}Sn . For the spin $7/2$ quadrupole moment in ^{111}Sn a ten fold reduction in uncertainty was achieved compared to the old measurements by J. Eberz *et al.* [76]. The quadrupole moment of the $7/2^+$ isomeric state in ^{113}Sn is larger than in ^{111}Sn , with a greater uncertainty due to the overlapping structure with its ground state. Measurements of the short-lived $7/2^+$ isomeric state in ^{115}Sn have been performed using PAC measurements in

³A comparison using different calculated EFGs are presented in appendix A.3.

Cadmium substrates by O. Echt *et al.* [180]. Considering that the EFG of Sn in Cd was determined with a very old semi-empirical model, it is worthwhile to verify if this value is still valid. Considering that the magnetic moments of the $5/2^+$ and $7/2^+$ isomers indicate a nearly perfect single particle behavior, we would expect the quadrupole moments to follow the seniority scheme, when filling respectively the $5/2^+$ and $7/2^+$ orbits. Thus, a re-evaluation of the ^{115}Sn $7/2^+$ isomeric state Q_s , using modern ab-initio based calculation methods for the EFG of Sn in Cd, is highly desired to verify such possible trend.

Table 7.5: Extracted B_{hf} -values (in MHz) from the measured hyperfine structures using the SP and PP transitions together with determined quadrupole moment Q_s compared to literature. The statistical uncertainty in Q_s is presented in round brackets and systematic uncertainty in square brackets. The B_{hf} -values shown in italics have been calculated from literature moments (see text for details).

Isotope	I^π	PP		SP	$Q_s^{Exp}[\text{b}]$	$Q_s^{lit}[\text{b}]$ [Ref.]
		$B_{hf}^{(l)}[\text{MHz}]$	$B_{hf}^{(u)}[\text{MHz}]$	$B_{hf}^{(u)}[\text{MHz}]$		
105	$5/2^+$	-38(4)	54(6)		0.055(7)[4]	
107	$5/2^+$	-124(3)	178(4)	90(6)	0.180(4)[12]	
109	$5/2^+$	-169(6)	243(8)	113(11)	0.245(8)[17]	0.218(7)[15] [77]
111	$7/2^+$	-88(4)	126(6)	37(6)	0.115(6)[9]	0.18(90) [76]
113m	$7/2^+$	-171(29)	246(42)	96(21)	0.250(42)[17]	
115	$7/2^+$					($\pm$)0.26(3) [180]
121	$3/2^+$	<i>39(4)</i>	<i>-55(6)</i>			-0.056(4)[4] [77]
121m	$11/2^-$	<i>64(6)</i>	<i>-92(8)</i>			-0.093(3)[7] [77]
123	$11/2^-$	0(4)	0(6)	1(9)	-0.001(9)[1]	-0.012(3)[1] [77]

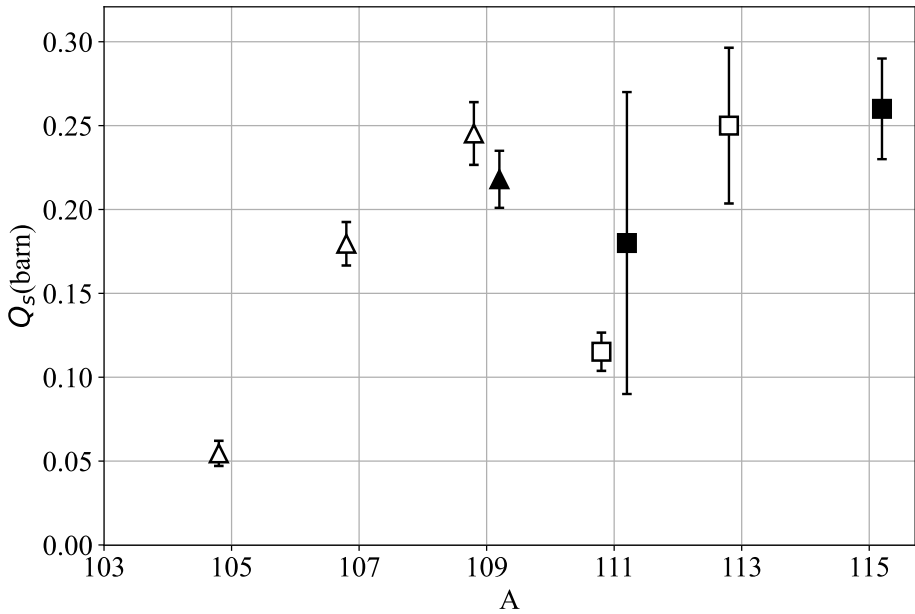


Figure 7.11: Measured spectroscopic quadrupole moments for the spin 5/2 isotopes $^{105,107,109}\text{Sn}$ with triangle makers, and for spin 7/2 nuclear states $^{111,113m,115m}\text{Sn}$ displayed with squares. The white markers indicate quadrupole moments from this work while black markers indicate measurements from literature, found in Ref. [77, 76, 180] for $^{109,111,115m}\text{Sn}$ respectively. The total uncertainty are indicated with error bars.

7.6 Isotope shifts

The drift corrected centroid frequencies ν^A of the fitted hyperfine structures with respect to the average value of ν^{112} (as seen earlier in Fig. 7.5) were used to determine the IS of the studied isotopes and isomers. IS of the SP and PP transitions are presented in Tab. 7.6 including values from the offline measurements, recalculated using ^{112}Sn as reference, for comparison. The offline and online measurements of both transitions are in good agreement within 2σ uncertainty for all measured isotopes.

Table 7.6: Measured IS (in MHz) using ^{112}Sn as reference obtained from the offline and online experiments in this work.

Isotope	I^π	PP		SP	
		$\delta\nu_{Online}^{A,112}$	$\delta\nu_{Offline}^{A,112}$	$\delta\nu_{Online}^{A,112}$	$\delta\nu_{Offline}^{A,112}$
104	0	-1726(11)			
105	$5/2^+$	-1523(8)			
106	0	-1204(7)		-902(10)	
107	$5/2^+$	-1017(7)		-784(7)	
108	0	-752(7)		-561(8)	
109	$5/2^+$	-592(8)		-450(9)	
110	0	-348(7)		-258(8)	
111	$7/2^+$	-246(9)		-177(7)	
112	0	0	0	0	0
113	$1/2^+$	138(19)		106(21)	
113m	$7/2^+$	85(16)		75(15)	
114	0	314(10)	342(43)	236(28)	249(28)
115	$1/2^+$	417(6)	425(41)	324(26)*	334(30)
116	0	655(10)	673(37)	491(10)	513(26)
117	$1/2^+$	755(7)	790(36)	591(24)*	599(29)
118	0	958(6)	986(35)	754(8)	749(26)
119	$1/2^+$	1048(10)	1078(34)	808(23)*	828(24)
120	0	1225(6)	1253(31)	950(24)*	971(23)
121	$3/2^+$	1284(28)			
122	0	1460(6)	1481(36)	1126(7)	1164(25)
123	$11/2^-$	1542(13)		1179(8)	
124	0	1678(6)	1715(37)	1277(8)	1313(29)

*These values have been calculated using IS from offline measurements with the average shift with respect to the neighbouring isotopes.

In order to verify the consistency of measured IS in different transitions the modified IS is compared in a KP as described in Sec. 3.5.1 using Eqn. 3.5.1. In Fig. 7.12 the modified IS of the PP transition $\Lambda_{(PP)}^{A,112}$ is plotted versus the modified IS of different transitions $\Lambda_{(JJ')}^{A,112}$ from [76, 85, 15] recalculated using ^{112}Sn as reference. The coefficients of the fitted linear function predicted by Eqn. 3.5.1 provide the ratio $F_\lambda^{(JJ')}/F_\lambda^{(PP)}$ and intercept C including the goodness-of-fit which are presented in Tab. 7.7. The value of $\chi_r^2 \leq 1$ indicates that the KPs are in better agreement than statistically expected from the given uncertainty in the measured IS. The ratio of $F_\lambda^{(JJ')}/F_\lambda^{(PP)}$ indicates that most transitions have a similar sensitivity to the nuclear charge radius, except for

the SP transition with a reduced FS factor by roughly 25% as predicted from the offline work presented in Paper I.

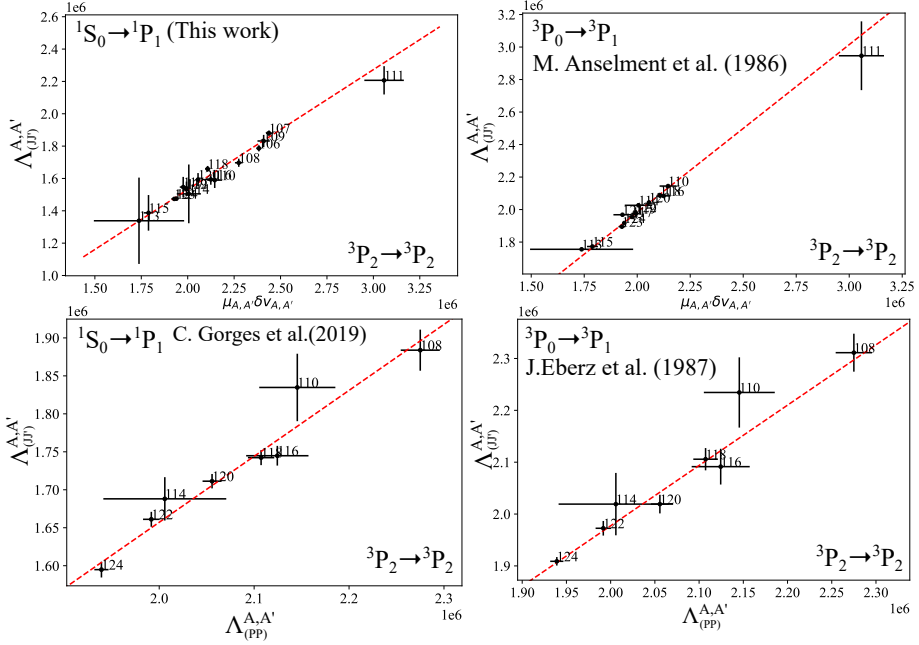


Figure 7.12: KP comparison with modified IS from the PP transition compared with IS of the SP transition and transitions from other work from Ref. [85, 15, 76].

Table 7.7: The presented coefficients were extracted from the linear trends presented in Fig. 7.12. Evaluated coefficients from comparing IS of different transitions with the PP transition, using Eqn. 3.5.1. The modified IS $\Lambda_{(JJ')}^{A,112}$ of a selected transition, indicated in the first column, are compared with $\Lambda_{(PP)}^{A,112}$.

Transition:	Ref.	$F_{\lambda}^{(JJ')}/F_{\lambda}^{(PP)}$	C [GHz u]	χ_r^2
$^3P_2 \rightarrow ^3P_2$ (PP)		1	0	-
$^1S_0 \rightarrow ^1P_1$ (SP)		0.737(32)	58(66)	1.017
$^1S_0 \rightarrow ^3P_1$	J.Eberz et al. [76]	0.942(50)	-231(100)	0.595
$^3P_0 \rightarrow ^3P_1$	M. Anselment et al. [85]	1.019(31)	-55(80)	0.267
	C. Gorges et al. [15]	1.163(72)	-349(144)	0.503

The FS and MS factors can be extracted from the IS of the stable tin isotopes with accurately determined charge radii using the approach described in Sec. 3.5.2. As an example, the modified IS of the PP transition is compared with the measured mean-square charge radii from [62] seen in Fig. 7.13. The extracted FS and MS factors for the different transitions using this procedure are provided in Tab. 7.8. These FS and MS factors are used to calculate the changes in mean-square charge radii from IS measured in this work including those from other transitions from [76, 85]. The reevaluated FS factor of the $5p^2\ ^3P_0 \rightarrow 5p6s\ ^1P_1$ transition further confirms the observation in both references [62, 107] with a roughly 50% lower FS factor compared to that used in previous literature found in [85, 84, 15].

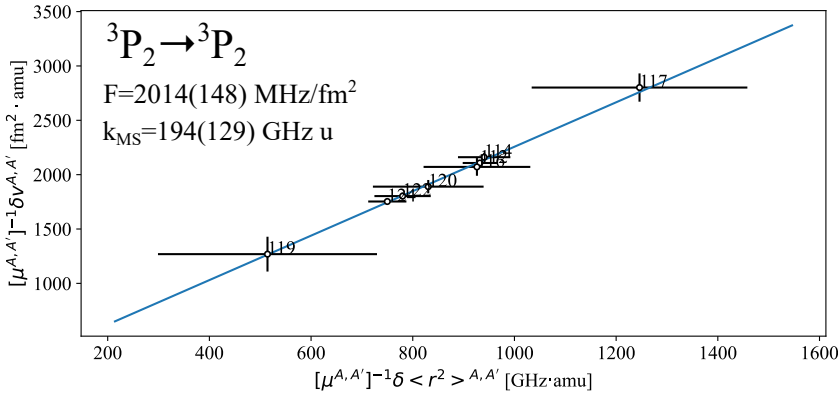


Figure 7.13: KP approach comparing modified IS from the PP transition with mean-square charge radii from [62]. The intercept and slope of the fitted linear trend provide the FS and MS factors for the PP transition.

Table 7.8: Reevaluated FS and MS factors using the approach described in Sec. 3.5.2. The IS data from data from J.Eberz *et al.* and M.Anselment *et al.* have been reevaluated.

Transition:	Ref.	F_{λ} [MHz fm ⁻²]	k_{ms} [GHz u]
$^3P_2 \rightarrow ^3P_2$		2014(148)	194(129)
$^1S_0 \rightarrow ^1P_1$		1552(233)	135(184)
$^3P_0 \rightarrow ^3P_1$	M.Anselment <i>et al.</i> [85]	2043(113)	170(96)
$^1S_0 \rightarrow ^3P_1$	J.Eberz <i>et al.</i> [76]	1802(158)	81(134)

7.7 Nuclear charge radii

Using the FS and MS factors presented in Tab. 7.8 together with masses and IS, the mean-square charge-radii can be calculated using Eqn. 3.18. The calculated mean-square charge-radii are presented in Tab. 7.9, with statistical uncertainties within round brackets and systematic uncertainties from the FS and MS factors within square brackets. The weighted-mean from both transitions are presented in the table with errors not reduced below minimal sample error. Furthermore, the rms charge-radii are calculated using; $R_{\text{rms}} = \sqrt{\delta \langle r^2 \rangle^{A,112} + (R_{\text{ref}}^{112})^2}$ with $R_{\text{ref}}^{112} = 4.594(1)$ fm from Ref. [62]. In Fig. 7.14 the rms charge-radii from this work with a shaded band indicating the systematic error, is compared with reevaluated rms charge-radii from Ref. [62, 174, 181, 182] using the recalculated FS and MS factors presented in Tab. 7.8. In order to highlight the finer details the three-point average can be determined which is defined accordingly,

$$\Delta_r^{(3)} = \frac{1}{2}(R_{A+1} - 2R_A + R_{A-1}). \quad (7.1)$$

The three-point average is plotted in Fig. 7.15 without systematic uncertainties, revealing the odd-even staggering effect discussed further in Sec. 8.4.2. This highlights the excellent agreement between measurements from this work compared with the reevaluated charge-radii from literature as expected from the KP comparison 7.12. These are the first evaluated charge-radii below ^{108}Sn together with the measured isomeric $7/2^+$ state in ^{113}Sn which can clearly be distinguished in the odd-even staggering in Fig. 7.15.

Table 7.9: Mean-square charge radii and rms charge radii calculated from isotope shifts. The round brackets contain statistical uncertainties while the systematic uncertainties from the atomic factors and the uncertainty in the reference rms charge radius (using ^{112}Sn from Ref. [62]) are contained within square brackets.

Isotope	$\delta\langle r^2 \rangle_{(SP)}^{A,112} [\text{fm}^2]$	$\delta\langle r^2 \rangle_{(PP)}^{A,112} [\text{fm}^2]$	$\delta\langle r^2 \rangle_{mean}^{A,112} [\text{fm}^2]$	$R_{rms}^{mean} [\text{fm}]$
104		-0.791(6)[77]	-0.791(6)[77]	4.5071(6)[86]
105		-0.699(4)[68]	-0.699(4)[68]	4.5173(5)[75]
106	-0.538(6)[106]	-0.549(4)[55]	-0.547(4)[55]	4.5341(4)[61]
107	-0.469(4)[91]	-0.465(3)[46]	-0.465(3)[46]	4.5431(4)[51]
108	-0.333(5)[67]	-0.342(3)[35]	-0.340(3)[35]	4.5569(4)[39]
109	-0.269(6)[52]	-0.27(4)[27]	-0.270(4)[27]	4.5645(4)[31]
110	-0.152(5)[32]	-0.157(3)[16]	-0.156(3)[16]	4.5770(4)[21]
111	-0.107(5)[20]	-0.114(4)[10]	-0.113(4)[10]	4.5817(5)[15]
112	0	0	0	4.594(0)[1]
113	0.061(14)[14]	0.061(10)[7]	0.061(10)[7]	4.6006(10)[13]
113m	0.041(8)[12]	0.034(8)[6]	0.037(8)[6]	4.5979(9)[15]
114	0.138(18)[29]	0.141(5)[15]	0.141(5)[15]	4.6093(5)[19]
115	0.188(17)[42]	0.185(3)[21]	0.185(3)[21]	4.6141(3)[25]
116	0.290(6)[60]	0.295(5)[31]	0.294(5)[31]	4.6259(5)[35]
117	0.348(15)[73]	0.338(3)[37]	0.340(3)[37]	4.6308(4)[41]
118	0.446(5)[91]	0.432(3)[46]	0.435(3)[46]	4.6411(3)[50]
119	0.475(15)[100]	0.470(5)[51]	0.471(5)[51]	4.6450(5)[56]
120	0.560(15)[116]	0.551(3)[59]	0.553(3)[59]	4.6538(3)[64]
121		0.573(14)[64]	0.573(14)[64]	4.6560(15)[69]
122	0.662(5)[140]	0.654(3)[71]	0.656(3)[71]	4.6648(3)[77]
123	0.690(5)[149]	0.689(6)[76]	0.689(5)[76]	4.6684(5)[82]
124	0.747(5)[161]	0.750(3)[83]	0.749(3)[83]	4.6748(3)[89]

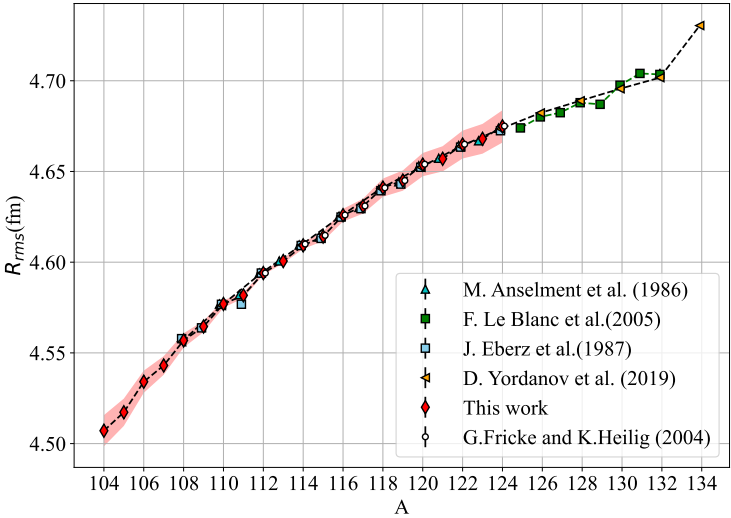


Figure 7.14: Tin charge radii from this work compared with recalculated charge radii from literature [181, 174, 182].

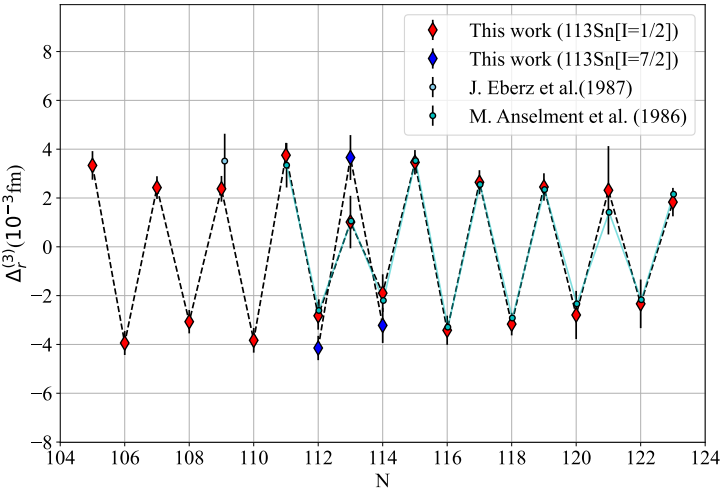


Figure 7.15: The three-point average (Eqn. 7.1) highlighting the odd-even staggering effect in good agreement with literature [181, 174].

Chapter 8

Discussion of experimental results

The results presented in the previous chapter extend our experimental knowledge of nuclear moments and charge radii along the tin isotopic chain down to ^{104}Sn , four neutrons away from the expected shell-closure at $N = 50$. Combined with experimental data reaching beyond $N = 82$ [15] these provide a unique testing ground for studying the evolution of nuclear properties from the neutron-deficient to the neutron-rich ends of the nuclear landscape.

There are three main questions discussed here: How does the ordering of the neutron $1g_{7/2}$ and $2d_{5/2}$ orbitals evolve when approaching ^{100}Sn ? What is the single particle nature of the neutron-deficient tin isotopes? Is an enhanced collectivity approaching $N = 50$ also observed in the charge radii? This chapter provides a discussion based on the presented results which shed a new light on these questions in particular.

In Sec. 8.1, the observed nuclear moments and spins of the odd- N neutron-deficient isotopes are described from a shell model perspective. With the first question in mind the measured g -factors and spins clarify the single particle nature of the ground- and isomeric states and reveal the ordering of the $1g_{7/2}$ and $2d_{5/2}$ neutron orbitals down to ^{105}Sn . The neutron occupancy in these two orbitals is discussed from the perspective of the measured magnetic dipole and spectroscopic quadrupole moments. These discussions open new questions regarding the continued nuclear structure evolution of $^{101,103}\text{Sn}$.

Sec. 8.4 presents precision measurements of nuclear charge radii along the tin

chain from $N = 84$ reaching down to $N = 54$. A characteristic parabolic trend in charge radii between the two shell-closures is investigated further, revealing the influence of a sub-shell closure at $N = 64$. The measured charge radii also reveal a new perspective of the evolution of collective properties along the tin isotopic chain. The deformation extracted from the measured charge radii is compared with the dynamic quadrupole deformation from measurements of $B(E2 : 0^+ \rightarrow 2^+)$ along the tin chain, revealing interesting similarities as well as clear discrepancies between the two approaches.

Finally, both the microscopic and collective properties of the tin chain are compared with state-of-the-art VS-IMSRG calculations using *ab-initio* based interactions. This comparison highlights the current progress and remaining challenges for reaching an *ab-initio* description of the tin isotopic chain and beyond.

8.1 A shell model perspective

8.1.1 Magnetic moments and the level ordering of tin

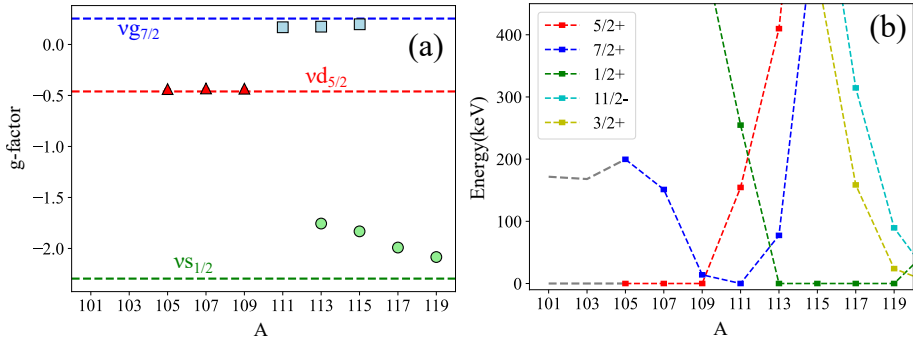


Figure 8.1: Experimental g-factors for the $2d_{5/2}$, $1g_{7/2}$ and $3s_{1/2}$ orbitals together with calculated effective g-factor for each orbital with $g_{eff} = 0.6g_{s.p.}$. (b) Energy level diagram for the nuclear states [176]. The ground-state level ordering has been firmly assigned in this work down to ^{105}Sn indicated with colors. The ordering below ^{105}Sn indicated with a grey dashed line remains unknown.

g-factors are particularly sensitive probes to the shell-model orbital(s) occupied by the unpaired neutron(s). Therefore g-factor measurements of the odd-N tin isotopes can be used for investigating the ordering of the $2d_{5/2}$ and $1g_{7/2}$

neutron orbitals when approaching ^{100}Sn . In this context, the evolution of g-factors from ^{119}Sn down to ^{105}Sn are presented in Fig. 8.1(a), together with the single particle energy levels in Fig. 8.1(b) using data from [176]. Owing to the firm spin assignment from this work the level ordering is now firmly established with a ground state spin of $5/2$ down to ^{105}Sn , as indicated with the colored trends in the Fig. 8.1(b).

The effective g-factors were calculated for a single neutron in the $2d_{5/2}$, $1g_{7/2}$ or $3s_{1/2}$ orbital respectively using Eqn. 2.12. A quenching factor of 0.6 was used to fit the constant g-factor trend for $^{105,107,109}\text{Sn}$ which is similar to the quenching used for other elements in the region, such as Cd and In [183, 109]. These calculated effective g-factors are presented with dotted lines in Fig. 8.1(a). A constant g-factor trend is observed for isotopes occupying the $2d_{5/2}$ orbital from ^{109}Sn down to ^{105}Sn , despite removing 4 neutrons from the nucleus.

Assuming a naive shell-model filling of a $2d_{5/2}$ orbital above a ^{100}Sn core, would imply a complete shell at ^{106}Sn . In this case a spin flip is expected at ^{107}Sn when the additional neutron is added in the $1g_{7/2}$ orbital. However, measurements suggests otherwise, where the $2d_{5/2}$ neutron orbital remains with a hole (thus unpaired neutron) in the $2d_{5/2}$ until ^{109}Sn . Calculations of the pairing energies presented by I. Darby *et al.* [22] reveal that $V^p(1g_{7/2}) = 1.4$ MeV and $V^p(2d_{5/2}) = 0.84$ MeV respectively. The energy difference for pair formation in the $1g_{7/2}$ and $2d_{5/2}$ of 0.56 MeV is significantly greater than the single particle energy difference between the two orbitals $\epsilon_{sp} < 0.200$ MeV, seen in Fig. 8.1. It is thus favorable for neutron pairs to occupy the $1g_{7/2}$ orbital. Consequently, the isotopes having a hole in the $2d_{5/2}$ orbitals are (according to determined spin and their constant magnetic moment) dominated by seniority-one configurations with $(1g_{7/2})^{2i} \otimes (2d_{5/2})^{2j+1}$ where i is the number of pairs in the $1g_{7/2}$ orbital and j is the number of neutron pairs in the $2d_{5/2}$ orbital.

While the next orbit is the $1g_{7/2}$, one would expect this to be the dominant ground state configuration from ^{111}Sn up to ^{115}Sn . However, only for ^{111}Sn the ground-state spin is found to be $7/2$, while this level appears as a low-lying isomeric state in $^{113,115}\text{Sn}$. The g-factor of these $7/2$ states is in very good agreement with that for an unpaired neutron in the $1g_{7/2}$ orbit. From ^{113}Sn onwards, the ground state spin is firmly assigned as $I = 1/2$. It is observed that these isotopes have a more mixed wave function partly due to the linear deviation from the constant g-factor trend, which has been confirmed by transfer reaction measurements [184]. The simple single particle trends are expected to break down as more neutrons are added within the shell as the residual interactions have an increased influence of the nuclear structure. The g-factor is thus further quenched for the $I = 1/2$ states due to the enhanced collectivity experienced when approaching midshell.

In summary, the measured g -factors and spins confirm a $5/2^+$ ground-state from ^{109}Sn down to ^{105}Sn . This can be explained by a seniority-1 configuration with an odd-neutron occupying the $2d_{5/2}$ orbital. A favoring pair energy in the $1g_{7/2}$, combined with the near-degeneracy of the $2d_{5/2}$ and $1g_{7/2}$ orbits, is a likely reason for the odd-neutron remaining in the $2d_{5/2}$ state up to ^{109}Sn . Further measurements are required to investigate if this simple picture indeed continues down to ^{101}Sn . The comparison to calculated magnetic moments using *ab-initio* calculations are presented in Sec. 8.5.2.

8.1.2 The quadrupole moment and neutron occupancy approaching ^{100}Sn

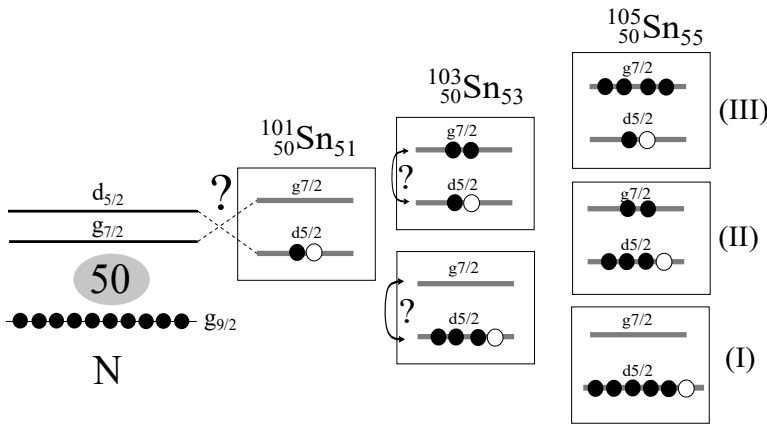


Figure 8.2: Possible configurations of the neutron shell-model orbits of $^{101}, ^{103}, ^{105}\text{Sn}$.

In the single-particle shell-model a single neutron added in an orbit above a closed shell will polarize the core which induces a negative quadrupole moment of the nucleus, see Sec. 2.5.2. This can be extended with the seniority scheme for added particles/holes in the shell-model orbit j which predict a linearly increasing trend with added neutrons in an j -orbit shell, with zero quadrupole moment when the orbital is half filled. As an example, the neutron $2d_{5/2}$ orbital is assumed above ^{100}Sn which has a maximum occupancy of 6 neutrons, see Fig. 8.2. In this case the seniority model predicts a linear trend crossing 0 at ^{103}Sn where the $2d_{5/2}$ orbital is half filled. Adding an additional neutron pair in the $2d_{5/2}$ orbital would result in a single neutron hole remaining in the $2d_{5/2}$ neutron orbital, configuration (I) in Fig. 8.2. In this configuration the

neutron hole would induce a positive quadrupole moment in ^{105}Sn . However, as discussed earlier neutron pairing in the $1g_{7/2}$ is favored. Assuming a neutron pair in the $1g_{7/2}$ neutron orbital for ^{105}Sn , configuration (II) in Fig. 8.2, would result in a half-filled $2d_{5/2}$ orbital and therefore zero quadrupole moment would be expected in this seniority model. Furthermore, for configuration (III) with a single neutron in the $2d_{5/2}$ orbital would result in a negative quadrupole moment.

In Fig. 8.3 the experimental quadrupole moments of the spin 5/2 and 7/2 tin isotopes and isomers are plotted from ^{115}Sn down to ^{105}Sn . The positive quadrupole moment observed for ^{105}Sn $Q_s(^{105}\text{Sn}) = 0.055(7)$ makes it clear that the measured value is in best agreement with configuration (I), one hole in the $2d_{5/2}$ neutron orbital. Assuming a sequential shell-model filling of the $2d_{5/2}$ neutron orbital above ^{100}Sn the seniority model can be used for calculating the single-particle quadrupole moment of the lightest tin isotopes using Eqn. 2.16, fixed to the experimental quadrupole moment of ^{105}Sn , with a zero crossing at ^{103}Sn . For $\nu = 1$, $j = 5/2$ together with the measured mean-square charge radii from Tab. 7.9 allows the extraction of the effective neutron charge from the slope of the linear trend. With this approach an effective neutron charge of $e_\nu^{eff} = 0.70(10)e$ was extracted, with uncertainty projected from the quadrupole moment of ^{105}Sn . This value is in excellent agreement with the effective neutron charge of $e_\nu^{eff} = 0.7e$ used for reproducing the quadrupole moment of ^{133}Sn , with a single neutron above the confirmed doubly-magic ^{132}Sn , presented in Ref. [17]. The measured quadrupole moment of ^{105}Sn is therefore in agreement with a simple sequential shell-model filling of the $2d_{5/2}$ neutron orbital above ^{100}Sn .

This observation together with the firm spin assignment and the constant g-factor trend suggest a continued trend with the $2d_{5/2}$ being the first orbital above ^{100}Sn . In this shell model seniority filling of the $2d_{5/2}$ neutron orbital above ^{100}Sn the quadrupole moment of ^{101}Sn is expected to be roughly equal but opposite to that of ^{105}Sn . However, this simple shell model behaviour is in contrast to the favored pairing in the $1g_{7/2}$ orbital. A configuration (II) for ^{105}Sn has suggested by I. Darby *et al.* [22] where a neutron pair remain in the $1g_{7/2}$ orbital. It is possible that the $1g_{7/2}$ neutron pair induces a positive quadrupole moment despite the $2d_{5/2}$ being half-filled. Measurements of the quadrupole moments and g-factors of ^{103}Sn and ^{101}Sn are therefore of particular interest in order to clarify this controversy.

As revealed from the g-factor measurements the odd-neutron remains in the $2d_{5/2}$ orbital as neutron pairs start filling the $2d_{5/2}$ for $^{107,109}\text{Sn}$. These isotopes exhibit a continued trend of increased quadrupole moment, greater than predicted assuming an extended seniority filling of the $2d_{5/2}$ orbital. This suggests that the simple single particle model breaks down with the added neutrons pairs, likely due to the additional coupling of neutrons in the $1g_{7/2}$ orbital inducing

additional correlations (and thus enhanced core polarization). As discussed by Togashi *et al.* [185], cross-shell excitations of protons from the $g_{9/2}$ orbital across $Z=50$ and the proton-neutron correlation with added neutrons in the $1g_{7/2}$ and $2d_{5/2}$ orbitals were essential in order to describe the enhanced collectivity of the neutron-deficient tin isotopes.

In the assumption of a generalized seniority (GS) filling of a $1g_{7/2} \otimes 2d_{5/2}$ configuration with $\Omega = 7$ (Eqn. 2.17), the two shells would be half-filled at $N=57$. It is clear that GS does not explain the trend for the quadrupole moment trend of $5/2$ states, however, the $7/2$ states are in better agreement with this model. The calculated quadrupole moment with GS filling was fixed to the quadrupole moment of ^{111}Sn ($I=7/2$) and the zero crossing at $N=57$, as presented with a black dashed line in Fig. 8.3. In order to reproduce the quadrupole moment of ^{111}Sn a significantly greater effective neutron charge of $e_{\nu}^{eff}=1.44(17)e$ was required for the odd-neutron in the $1g_{7/2}$ orbital. Additional core polarization effects are required to explain the enhanced quadrupole moment with added neutrons towards midshell.

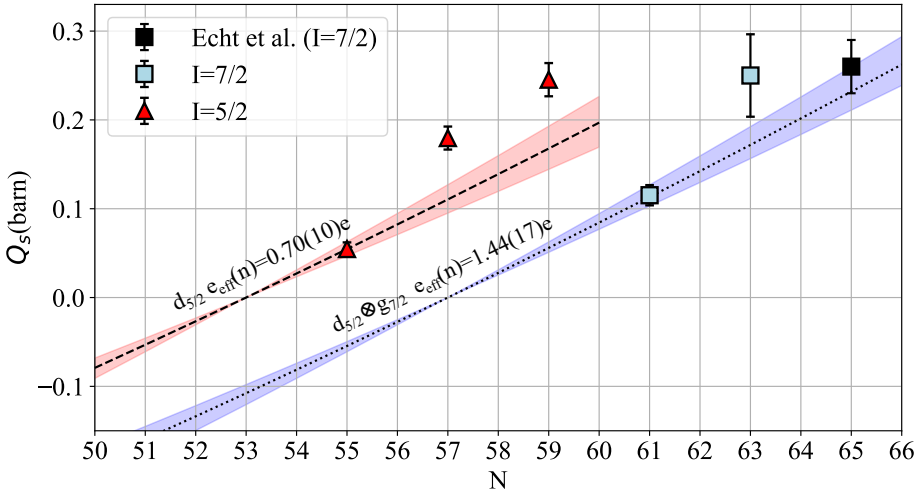


Figure 8.3: Quadrupole moment of neutron-deficient tin isotopes. Two trends have been fitted to the data: The dashed red trend is calculated using the seniority scheme assuming shell-model filling of the $2d_{5/2}$ neutron orbital, where the line is fitted through ^{105}Sn and crossing zero at the half-filled orbit. The second trend indicated with black dashed lines is calculated from the generalized seniority filling of the $1g_{7/2} \otimes 2d_{5/2}$ configuration with $\Omega = 7$, crossing the mid-point filling at $N=57$, see Eqn. 2.17.

8.2 Comparing Sn with Cd

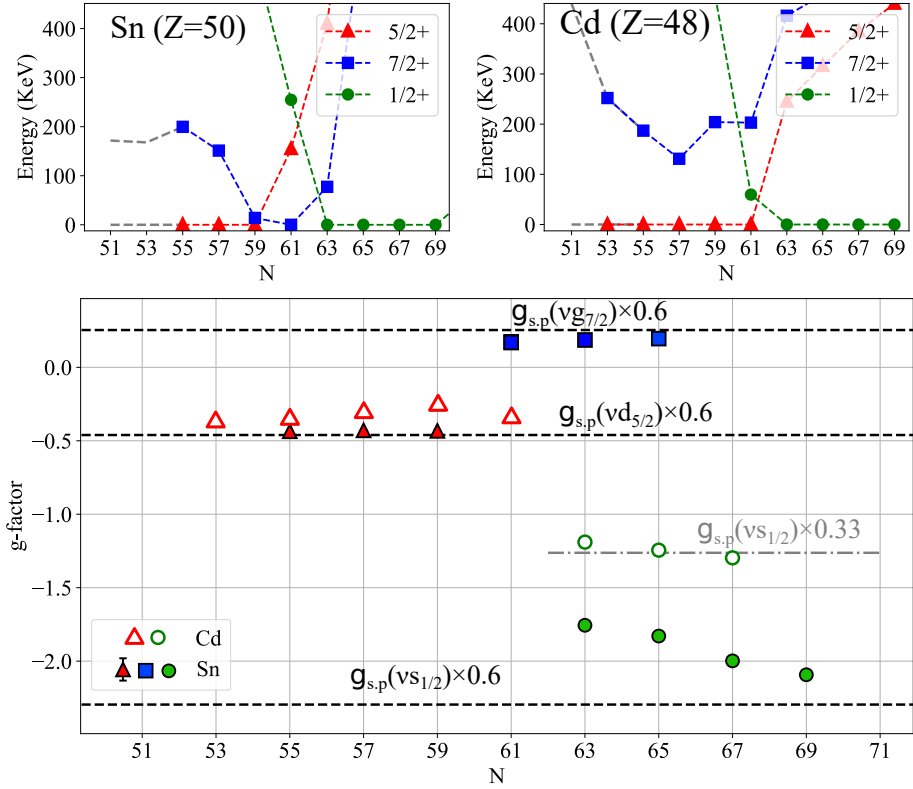


Figure 8.4: At the top are two energy level diagrams for the lowest lying nuclear states in Sn ($Z=50$) and Cd ($Z=48$), using data from Ref. [176]. The ground-state level ordering has been firmly assigned in this work down to ^{105}Sn ($N=55$). The grey dashed lines indicate unknown spin assignment. The figure below shows the experimental g-factors for the $5/2^+$, $1/2^+$ and $7/2^+$ states in the Cd and Sn isotopes, along with the effective g-factors for the $2d_{5/2}$, $1g_{7/2}$ and $3s_{1/2}$ neutron orbits with different quenching factors as indicated in the figure.

The level ordering and nuclear moments of Sn ($Z = 50$) can be compared with Cd ($Z = 48$) to investigate the influence of two proton holes in the $1g_{9/2}$ orbital on the level structure and g-factors. In Fig. 8.4 the level ordering of lowest lying states in neutron-deficient Sn and Cd are presented together with the g-factors for each orbital on the level structure and g-factors. There is a clear resemblance between the orbital arrangement for the two elements with the

main difference being the $1g_{7/2}$ orbital ordering. While a $7/2^+$ ground state is observed at $N = 61$ in tin, a $5/2^+$ ground state is observed for Cd, which is also revealed from the measured g-factors at $N = 61$. While the $7/2^+$ and $5/2^+$ levels differ by at most 200 keV in all isotopes of Cd and Sn, the two levels become near-degenerate in ^{109}Sn . The lowering of the $1g_{7/2}$ neutron orbital in tin is attributed to the attractive monopole tensor force between the added protons in the $1g_{9/2}$ and neutrons in the $1g_{7/2}$ orbital [186]. When both the proton $1g_{9/2}$ and neutron $1g_{7/2}$ are fully filled then the tensor force reduces to zero which is evident from the rapidly increasing energy of the $7/2^+$ state above $N = 64$.

While the g-factors at $N = 55$ are nearly the same for both elements the g-factor trends diverge with added neutrons in the shell. This is particularly noticeable for the $1/2^+$ states where a significantly greater quenching is required to account for the residual interactions in Cd. Here, the single particle g-factor for the $1/2^+$ states in Cd require a quenching of 0.33. This is likely caused by the enhanced collective effects resulting from the coupling with the proton holes in the $1g_{9/2}$ orbital.

The trend of measured spectroscopic quadrupole moments for isotopes with an odd-neutron occupying the $2d_{5/2}$ orbital are presented for both Sn and Cd isotopes in Fig. 8.5. An enhanced quadrupole moment in Cd compared to Sn is seen. While an effective neutron charge of 0.70(10)e is required in order to explain the single particle quadrupole moment of ^{105}Sn , an effective charge of 2.20(11)e is extracted assuming seniority filling of the $2d_{5/2}$ orbital fitted through $N = 53, 55$ for Cd. The quadrupole moment trend in Cd for the $5/2$ states is discussed in Ref. [187]. In this discussion an occupancy of 3 neutron in the $2d_{5/2}$ orbital is suggested at $N = 55$, which was based on the zero quadrupole moment observed in ^{101}Cd ($N = 55$). However, the proton holes in Cd are expected to have a constant positive core deformation caused by the missing proton pair in the $1g_{9/2}$ orbital, as seen when comparing the $2d_{3/2}$ and $1h_{11/2}$ orbitals in Cd and Sn [77]. Interestingly, the opposite is seen here, where a smaller quadrupole moment is seen in Cd compared to Sn at the crossover point at $N=55$. These questions further motivates the continued spectroscopic measurements below ^{105}Sn .

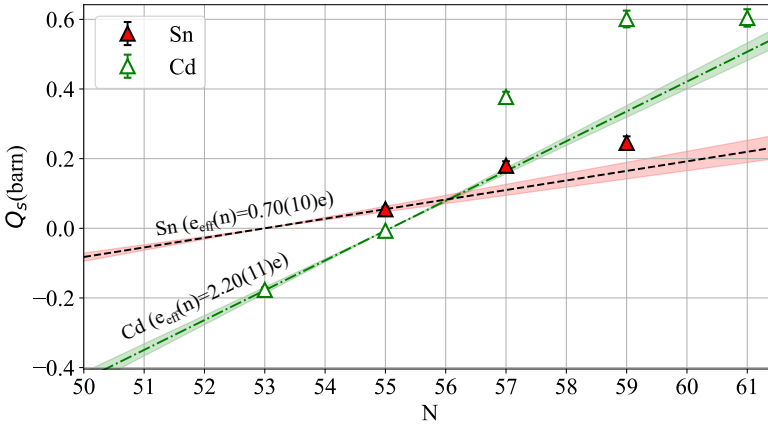


Figure 8.5: Spectroscopic quadrupole moment of odd-even Sn isotopes occupying the $2d_{5/2}$ orbital (in red) and for Cd isotopes indicated in hollow green triangles taken from Ref. [187].

8.3 Conclusions from a shell model perspective

This discussion reveals that the single particle shell model is limited in explaining the ground state properties of Cd and Sn isotopes. However, the combined insight from measured g-factors and firm spin assignment, is consistent with a seniority-1 configuration with an unpaired neutron in the $2d_{5/2}$ orbital from ^{109}Sn down to ^{105}Sn . The unpaired neutron remains in the $2d_{5/2}$ orbital due to the favored neutron pairing in the $1g_{7/2}$ orbital, explaining the spin 5/2 ground-state in $^{107,109}\text{Sn}$. Interestingly, the quadrupole moment of ^{105}Sn is in excellent agreement assuming seniority filling with a single hole in the $2d_{5/2}$ orbital above ^{100}Sn with an effective charge similar to a single neutron above the doubly-magic ^{132}Sn . This observation could be an indication of a strong shell-closure at ^{100}Sn assuming shell-model filling of the $2d_{5/2}$ as the ground state arrangement above ^{100}Sn . However, it is not possible to rule out that the observed quadrupole moment is a result of core polarization induced by the coupling of neutrons in the $1g_{7/2}$ orbital with the protons excited from the $1g_{9/2}$ orbital across $Z = 50$. This process is the likely reason for the enhanced quadrupole moment observed for $^{107,109}\text{Sn}$ which point to an increased collectivity occurring here. Spectroscopic measurements of ^{103}Sn and ^{101}Sn are of particular interest to investigate continuation of the suggested shell-model trend above ^{100}Sn .

8.4 Trends in nuclear charge radii

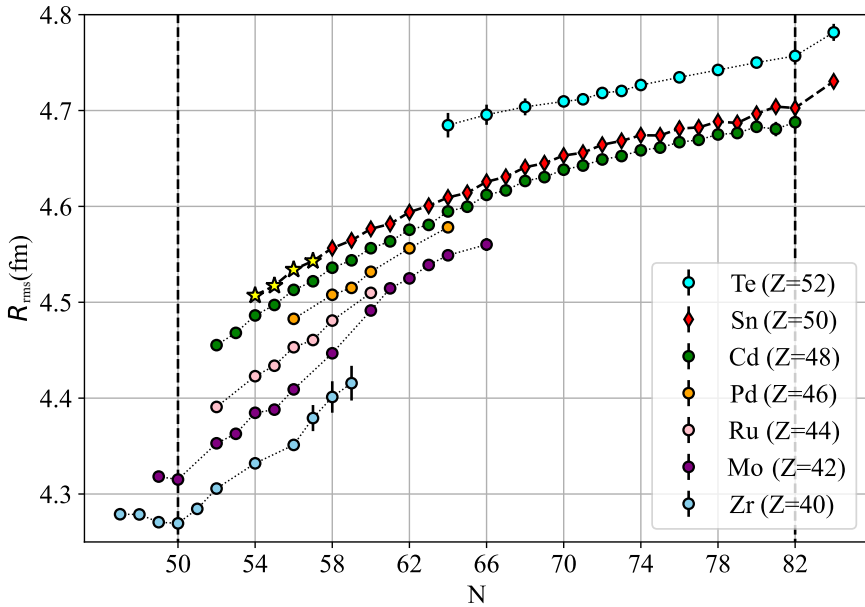


Figure 8.6: Rms charge radii (R_{rms}) of even- Z elements as a function of neutron number. The newly measured tin radii are highlighted with stars with literature data taken from Ref. [188, 15, 84].

The absolute rms charge radii of tin isotopes are presented together with isotope chains of nearby elements with even proton number, ranging from Te ($Z = 52$) to Zr ($Z = 40$) in Fig. 8.6. The new measurements from this work are highlighted with stars along the tin isotope chain reaching down to the neutron-deficient ^{104}Sn . Charge radii in this mass region follow characteristic parabolic trends between the shell closures at $N = 50, 82$. Distinct breaking points are observed in these trends when crossing the magic numbers. These breaking points are telltale signs of strong shell-closures, this was observed when adding a neutron pair to ^{132}Sn which indicated the doubly-magic shell-closure at $N = 82$, $Z = 50$ [15]. On the other side another clear kink in radii is observed when crossing $N = 50$ as seen in both molybdenum and zirconium isotopic chains, pointing to the occurrence of the $N = 50$ shell-closure [189, 190]. Recent spectroscopic measurements of neutron-deficient Ag ($Z = 47$) reveal that the $N = 50$ shell closure is maintained with only three holes in the $g_{9/2}$ proton orbital [191]. Furthermore, the proton shell-closure at $Z = 50$, is visible from the sudden

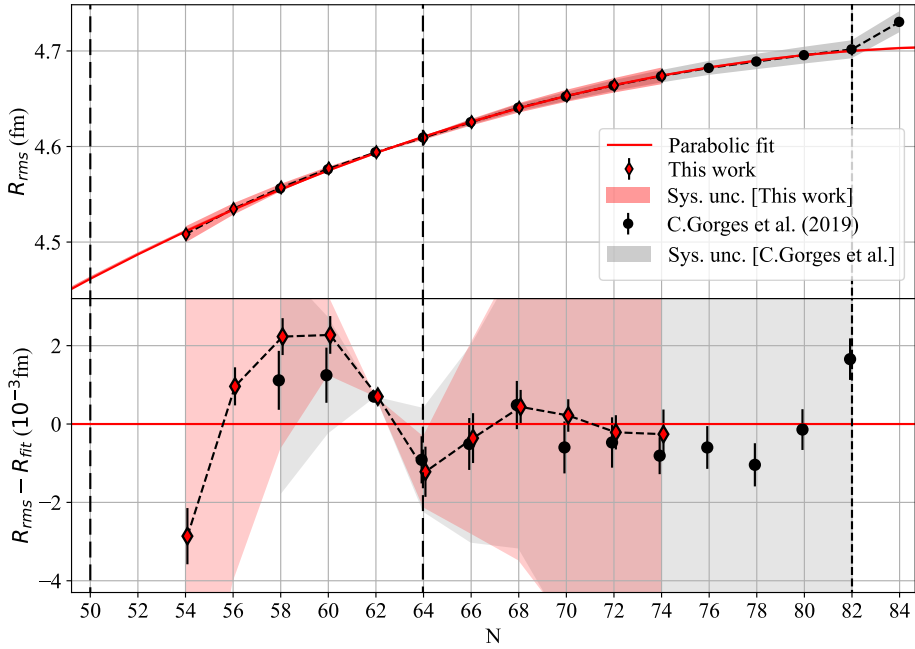


Figure 8.7: Rms charge radii of even-even tin isotopes from this work together with recent work from [15, 76]. Charge radii for isotopes below ^{132}Sn have been fitted using a parabolic function and the residuals of the fit are used for enhancing the deviation from the parabolic trend, revealing a sub-shell closure at $N = 64$ and a strong deviation at $N = 54$. Only statistical uncertainties are included when fitting of the parabolic function (not including ^{112}Sn).

jump in radii from Sn to Te when adding two protons beyond $Z = 50$. This highlights the sensitivity of measured charge radii to the nuclear shell structure.

In order to reveal the finer details of trend in charge radii, a quadratic function was fitted to the trend of the charge radii of even-even tin isotopes up to ^{132}Sn , as seen in Fig. 8.7. At first glance the fitted parabola shows good agreement with the measured charge radii. However, by looking at the deviation of the data from the parabolic trend, interesting details emerge from the high-precision measurements. The expected parabolic trend is broken at the sub-shell closure at $N = 64$ followed by a bump towards lower N , which continues in a steep decline towards ^{104}Sn , far steeper than expected according to the parabolic fitted function. This is consistent with the observations from the quadrupole moments, which pointed to the single-particle behavior of ^{105}Sn , and enhanced core polarization from ^{107}Sn onwards when adding neutrons in the $1g_{7/2}$ orbital.

Evidence collected from electron scattering, muonic x-ray and laser spectroscopic measurements have also hinted to a sudden jump in the charge distribution when adding neutrons beyond ^{114}Sn [192, 193, 85, 76], which was interpreted as due to the sub-shell filling of 14 neutrons in the $[d5/2, g7/2]$ orbitals. A similar sub-shell effect was first observed at $Z = 64$ from gamma and beta spectroscopic measurements of ^{146}Gd ($Z = 64$, $N = 82$) by P. Kleinheinz *et al.* [194]. This shell closure was also observed from a measured jump in charge radius when adding a proton beyond $Z = 64$ in Tb ($Z = 65$) [195]. Compared to the major magic shell closures predicted by the nuclear shell model, the $Z = 64$ shell closure is significantly weaker and is therefore called a *submagic* number [196].

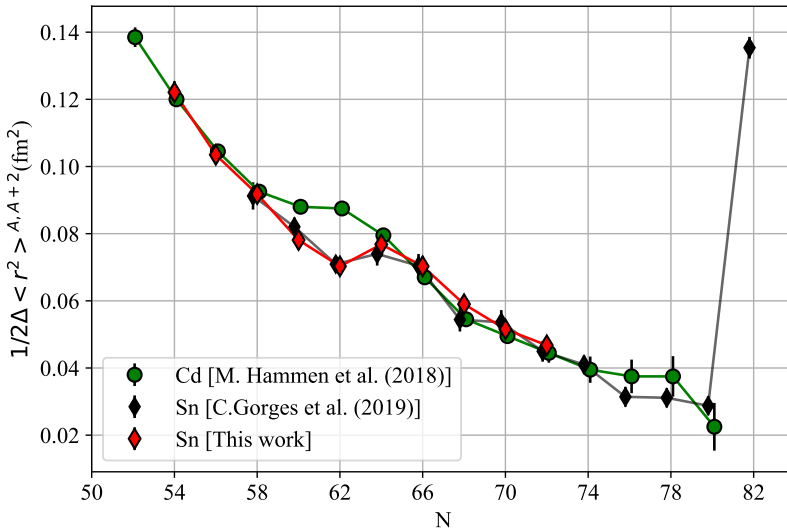


Figure 8.8: Brix-Kopfermann diagram of differential charge radii of even-even tin isotopes, compared to literature [15]. Charge radii of even-even Cd isotopes are added for comparison from Ref. [197].

Small changes in charge radii occurring along the isotopic chains are often studied using the differential mean-square charge radii;

$$\frac{1}{2}\Delta\langle r^2 \rangle^{A,A+2} = \frac{1}{2}(\langle r^2 \rangle^{A+2} - \langle r^2 \rangle^A). \quad (8.1)$$

This represents the change in radius with added neutron pairs to the nucleus and is often referred to as a Brix-Kopfermann diagram [198]. Since the even-even

isotopes are directly compared to each other, the systematic effects from the atomic factors are reduced and effects related to odd-even staggering are ignored. The Brix-Kopfermann diagram of the even-even tin isotopes are presented in Fig. 8.8, including radii from C. Gorges *et al.* [15] and compared with even-even Cd radii [197].

The diagram reveals the similarities between the two isotopic chains while also highlighting a discrepancy around $N = 62$. The kink in Sn radii when adding a neutron pair to the doubly-magic ^{132}Sn ($N = 82$) is seen in the figure as a sudden increase of the differential radii. The sub-shell closure at $N = 64$ in tin is also revealed, which is roughly 20 times weaker compared with the doubly-magic shell closure seen at $N = 82$. This explains why sub-shell effects are often missed when studying the changes in charge radii, instead of differential radii, and it highlights the importance of high-precision measurements of charge radii, which are essential for revealing these subtle features in the nuclear landscape.

While measurements of Cd radii has also revealed a feature near the $N = 64$ sub-shell closure [199], it is much less evident compared to tin as seen in Fig. 8.8. It appears that removing two protons from the $Z = 50$ closed shell leads to a distinctly different signature in the change in radii when transitioning the sub-shell closure. So far, many other attempts have been made to describe the subtle features in radii along the tin chain using a variety of different approaches such as semi-empirical droplet models and shell-model calculations using phenomenological interactions, with limited success [85, 200, 201, 15]. It is therefore clear this is a remaining challenge for testing the precision of nuclear theory.

8.4.1 Dynamic deformation

As described in Sec. 2.6, nuclear deformation contributes to the redistribution of nuclear charge in the surface region of the nucleus. Hence, the effects of deformation can be investigated if the spherical equivalent radius is known. In the case of even-even $I = 0$ isotopes there is no measurable static quadrupole deformation of the charge distribution. However, excitations to vibrational or rotational collective states have been studied using various gamma spectroscopy techniques [202]. These observations suggests that the collective excitation in tin are dominated by harmonic vibrations of the nuclear surface as expected for semi-magic nuclei near shell-closures [28].

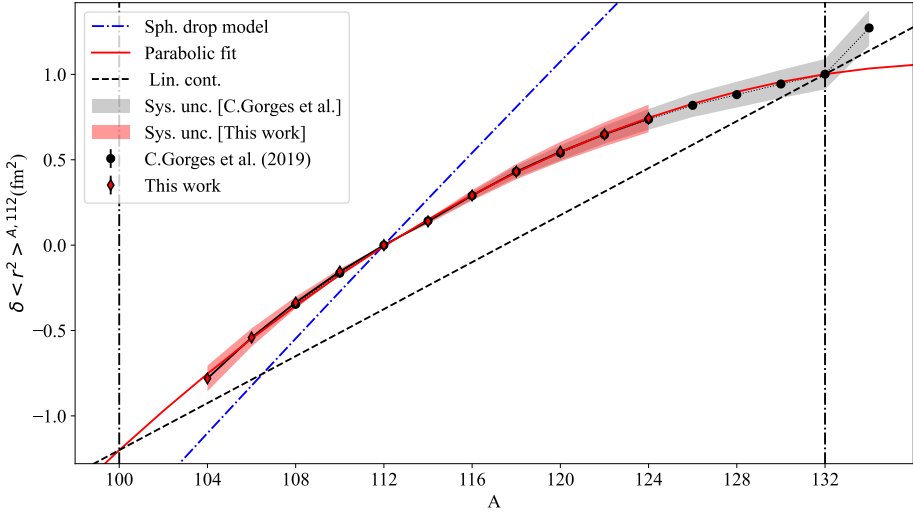


Figure 8.9: Differences in mean-square charge radii of even-even tin isotopes using ^{112}Sn as reference. The data has been fitted using Eqn. 8.2. The linear component which is fixed to cross the parabola in ^{100}Sn and ^{132}Sn , is compared with the spherical droplet model using $r_0 = 1.2 \text{ fm}$ (Eqn. 2.18).

The mean-square charge radius calculated using the spherical droplet model is to a very good approximation linear, as seen in Fig. 8.9. This allows us to construct a model describing the trend in radii as a combination of a linear component corresponding to the spherical contribution and parabolic term from the collective deformation accordingly,

$$\delta \langle r^2 \rangle_{par.}^{A, A'} = \underbrace{a(N - 82) + c}_{\text{Spherical cont.}} + \underbrace{b(N - 50)(N - 82)}_{\text{Deformation cont.}}. \quad (8.2)$$

Here, the linear term accounts for the spherical contribution which is fixed at the confirmed spherical doubly-magic ^{132}Sn used as a pivot point for this model such that $c = \langle r^2 \rangle^{132}$. The quadratic term describes the radial contribution resulting from collective vibrations of the nucleus, assuming perfectly rigid shell closures at $N = 50$ and 82 . The mean-square charge radii between the shell-closures were fitted using Eqn. 8.2, seen as the red curve in Fig. 8.9. The coefficients of the linear term: $a = 0.0694(10) \text{ fm}^2$ and quadratic term $b = -0.00153(6) \text{ fm}^2$ were extracted from the fitted model. The slope of the linear trend, describing the spherical contribution, is in good agreement with the slope predicted by semi-empirical spherical droplet models [76, 203, 11]. The influence of the dynamic deformation on the radius can be directly investigated by subtracting the spherical contribution (linear term) from the experimentally measured radii

using Eqn. 2.23. In Fig. 8.10 the dynamic deformation in terms of the summed contribution of the $\sqrt{\sum \langle \beta_i^2 \rangle}$ have been extracted from the experimental charge radii. As a comparison the dynamic quadrupole deformation $\langle \beta_2^2 \rangle^{1/2}$ have been calculated from $B(E2)$ measurements [78, 79, 21, 19, 20, 80, 81, 82, 83] using Eqn. 2.25.

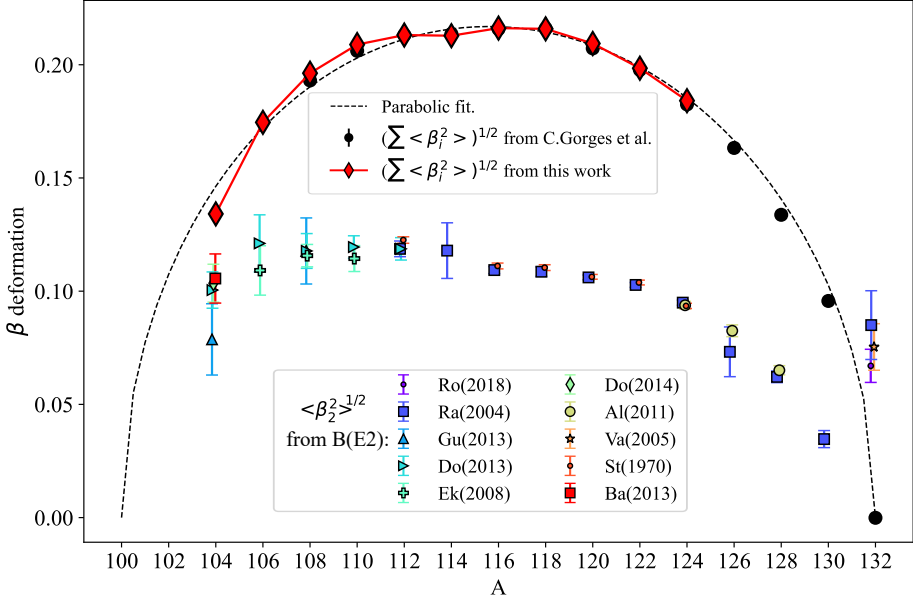


Figure 8.10: The deformation parameter β calculated from mean-charge radii from this work (red) and from Ref. [15] (black) compared with $\langle \beta_2^2 \rangle^{1/2}$ calculated from $B(E2)$ transition probabilities [78, 79, 21, 19, 20, 80, 81, 82, 83].

While both approaches indicate reduced deformation approaching ^{132}Sn , $B(E2)$ measurements suggest an enhanced dynamic quadrupole deformation towards ^{100}Sn . Many previous attempts have failed to describe this asymmetry, where a smooth parabolic trend in collective deformation was expected between shell-closures as presented earlier in Sec. 2.7. In contrast to the $B(E2)$ measurements, the deformation extracted from charge radii indicate a clearly decreasing trend towards ^{100}Sn , closely following the parabolic trend. However, as noted earlier the parabolic trend is broken at $N = 64$ by the sub-shell closure followed by a small 'bump' ending in a steep decline down to ^{104}Sn which is also seen in Fig. 8.10.

As suggested by Morales *et al.* [204] the asymmetric trend observed in the $B(E2)$ along the tin chain can be interpreted from a generalized seniority

filling of the shell model orbitals. In this interpretation the collectivity of the neutron-deficient tin isotopes below $N = 64$ is driven by the neutron occupancy in the $2d_{5/2}$ and the $1g_{7/2}$ neutron orbitals which explain the enhanced collectivity occurring when the sub-shell is half-filled, around ^{108}Sn . Once neutrons are added beyond $N = 64$ the collectivity is driven by the filling of the $3_{1/2}$, $2h_{11/2}$, $2d_{3/2}$ neutron orbitals between $N = 64$ and $N = 82$ which results in the observed double bump trend [205]. The asymmetric trend has also been investigated using large scale Monte-Carlo shell model calculations using a valence model space with a ^{80}Zr core (see Fig. 2.2), thus allowing for excitations of protons across $Z = 50$ [185]. These calculations well reproduced the asymmetric trend with the enhanced $B(E2)$ observed for the neutron-deficient tin isotopes and highlighted the importance of protons excitation across the $Z = 50$ shell gap. Such proton excitations are favored in the neutron-deficient tin isotopes, because opening up the proton $1g_{9/2}$ space allows for enhanced strong proton-neutron interactions with the neutrons occupying the $1g_{7/2}$ orbital. As these orbits are spin-flip partners, such interactions are strong, due to the tensor force as described earlier in Sec. 8.2. The bump observed below $N=64$ from the measured charge radii together with the enhanced quadrupole moment of ^{107}Sn and ^{109}Sn with added neutrons in the $1g_{7/2}$ orbital, provide an additional perspective of the influence of proton excitations across $Z=50$.

There is a notable discrepancy between β extracted from the mean-square charge radii and from $B(E2)$ measurements in the tin region, which has been a long standing question for many decades [206, 172, 207, 76]. While the β extracted from charge radii include all forms of deformation of the charge distribution, such as higher harmonic vibrations and changes in the skin thickness etc. The measured $B(E2)$ values only probe the excitation probability to quadrupole vibrations of its surface. However, higher-order vibrational modes such as octupole, hexadecapole etc. are expected to account for only a minor contribution of less than 5% of the dominant quadrupole vibrations [208].

It is important to acknowledge the strong model dependence of the approach used [209], where it is assumed that the dynamic deformation contribution can be easily decoupled from the total charge distribution. However, electron scattering measurements indicate a complex charge distribution in tin nuclei where the simple Fermi charge distribution breaks down [192, 210]. These measurements have revealed a reduced charge density at the center of tin nuclei (known as a core hole), as well as a slight variation in the skin thickness of their charge distribution (shown in Fig. 2.8) with added neutrons.

Measurements of the higher-order radial moments could reveal a more detailed picture of the charge distribution. However, such measurements have only been performed in the range of stable isotopes from ^{124}Sn to ^{112}Sn where the parabolic trend is almost invisible. A more sophisticated description of the

nuclear charge distribution is likely required in order to accurately decouple the contribution of the different deformation contributions as discussed by Papoulia et al. [211]. This highlights the need for measurements of the higher-order radial moments in short-lived tin isotopes for clarifying the influence of deformation of the nuclear charge distribution. Such measurements could potentially be realized by performing electron scattering on radioactive nuclei in storage rings [212].

8.4.2 Odd-even staggering

The three-point average (Eqn. 7.1) is used to highlight the odd-even staggering effect along the tin isotopic chain, seen in Fig. 8.11, compared with the three-point average of Cd radii from [197]. The staggering effect presents itself as an increase in the charge radius for isotopes containing an even number of nucleons compared to its neighbouring isotopes with an odd number of nucleons. The two isotope chains show remarkable similarities with only minor discrepancies. Notably, a reduced odd-even staggering is observed at $N=63$ compared to Cd. This shows the suppressed odd-even staggering occurring for the spin $1/2$ ground-state in ^{113}Sn compared to the spin $7/2$ isomeric state as clearly seen in Fig. 7.15, providing an interesting example of the sensitivity to the single particle behaviour from measured charge radii.

Different phenomenological models have been developed over the years to describe these features. Such as the odd proton or neutron blocking of ground-state quadrupole correlations or in terms of deformation, discussed in more detail in the references [213, 214, 215]. More recently, *ab-initio* based calculations methods accounting for N-body correlations show a remarkable agreement with the odd-even staggering effect observed in measured radii of copper isotopes [164]. A comparison with these modern *ab-initio* calculations are presented in the following section.

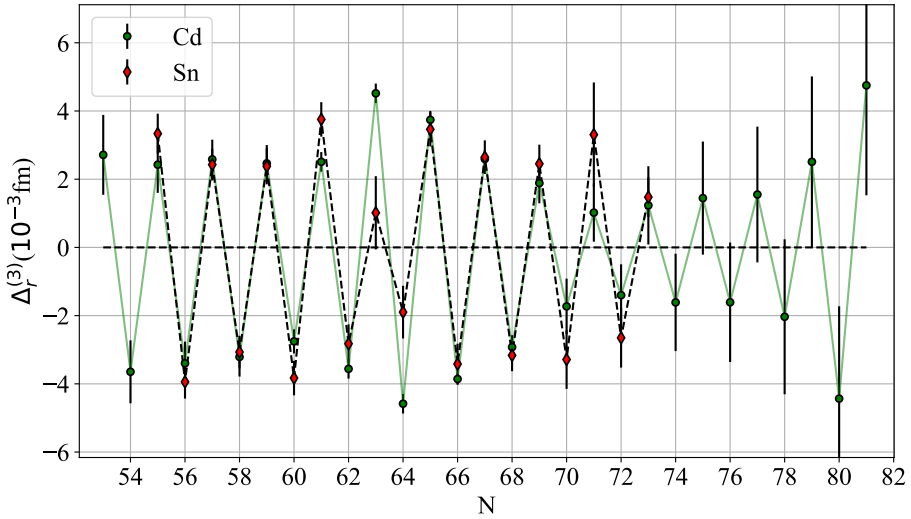


Figure 8.11: Three-point average of the nuclear charge radii of Cd (Ref. [197]) and Sn chain.

8.5 *Ab-initio* description of tin nuclei

The motivation to reach a QCD based description of the nucleus has driven modern *ab-initio* based calculation methods to describe properties of increasingly more complex nuclei. Due to the limitations in computational power one often uses doubly-magic nuclei as the inactive core for describing the greater regions of the nuclear landscape. As described earlier, ^{100}Sn is one of the last fundamental stepping stones in the nuclear landscape predicted by the nuclear shell model. Therefore the data provided in this work are of particular value for benchmarking the progress of *ab-initio* calculations using ^{100}Sn as a truncation point.

Two nuclear interactions EM1.8/2.0 and N2LO_g founded on the fundamental symmetries of QCD were employed in this work together with the VS-IMSRG framework for calculating the energies, nuclear moments and charge radii of the tin isotopes. Using the VS-IMSRG approach the effective Hamiltonian could be derived from the free-space interactions for selected valence spaces using either a ^{100}Sn or a ^{80}Zr core, see Fig. 2.2. Here, the effective Hamiltonians were calculated from a 13 harmonic oscillator (HO) major-shell space ($\text{emax}=12$) with frequency $\hbar\omega = 16$ MeV. Due to the memory limitation, an additional truncation $E_{3\text{max}}$ was introduced as the sum of the three-body HO quanta. In the presented calculation, $E_{3\text{max}}=16$ (24) [55] was used for the ^{100}Sn (^{80}Zr) core calculations. The diagonalizations of the effective Hamiltonians were performed using shell model code (KSHELL) for extracting the energy eigenvalues. The effective operators M1, E2 were derived using IMSRG++ code [56] which allowed the calculations of nuclear moments and charge radii in this work. The calculations presented in this section are all preliminary.

8.5.1 Ground-state level ordering

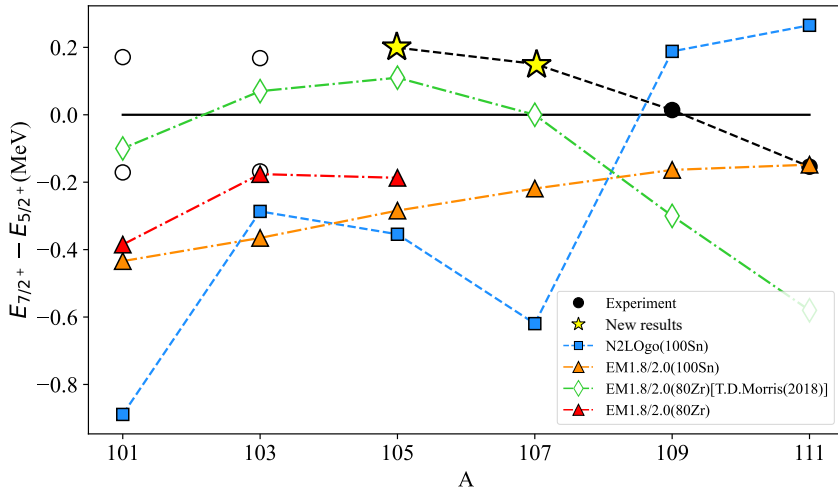


Figure 8.12: Calculated energy differences of the $7/2^+$ and $5/2^+$ states of odd tin isotopes using VS-IMSRG calculations with different interaction and core. Old calculations from T.D. Morris *et al.* [27] are presented with hollow green diamonds.

The VS-IMSRG framework with the two *ab-initio* based interactions were utilized to investigate the ground-state level ordering of the $2d_{5/2}$, $1g_{7/2}$ neutron orbitals from ^{101}Sn up to ^{111}Sn . In Fig. 8.12 the calculated energy difference and ordering of the lowest lying states are compared with experiment indicated in black with stars highlighting the new spin assignments in this work. The EM1.8/2.0 calculations using a ^{100}Sn core are indicated in orange and in red when using a ^{80}Zr core. Earlier work from T.D Morris *et al.*, using the same EM1.8/2.0 interaction calculated using VS-IMSRG with a ^{80}Zr core are shown with hollow green diamonds. Finally, the N2LOgo calculations using a ^{100}Sn core are presented in blue. While all calculations correctly predict the close energy spacing between the two orbitals < 1 MeV the precision and sign varies between the different interactions and valence space used.

Firstly, looking at the VS-IMSRG calculations using the EM1.8/2.0 interaction with a ^{100}Sn core (orange triangles). These calculations consistently favour a spin $7/2$ ground state with reduced energy spacing for increasing neutron number up to ^{111}Sn . While the level ordering and energy difference of ^{111}Sn is well predicted the change in level ordering to the $5/2$ state is not in agreement. Calculations using the larger valence space of a ^{80}Zr core (red triangles) were

performed up to ^{105}Sn . At ^{101}Sn both calculations are in agreement. While these calculations had little influence on the level spacing for ^{101}Sn they predict a smaller level spacing in better agreement with experiment for $^{103,105}\text{Sn}$, with a better prediction of the trend of the energy levels. Regardless, the ordering of the orbitals in ^{105}Sn remain reversed as compared to experiment, indicating something is still missing in these calculations. While in the earlier work by Morris *et al.* [27] (green diamonds) also using the ^{80}Zr core with the same valence space and interaction, there is a clear difference between between the two calculations. This difference is likely attributed to the choice of the reference state. In the previous work the Hartree-Fock (HF) reference state were calculated with the fixed level ordering predicted by the conventional nuclear shell model. However, in the current calculation, the level ordering can be changed during the HF iteration self-consistently, such that the HF energy can be minimized properly. Although the reference state choice should vanish when the IMSRG evolution is done without any approximation, the many-body truncation is inevitable in practice. Since the choice can propagate to the error of the IMSRG calculation, this highlights that spin assignment is still a major challenge for nuclear *ab-initio* methods, in particular in cases with nearly degenerate energy levels.

The N2LO_g interaction (blue squares) on the other hand favor a $5/2$ ground-state above ^{107}Sn but show a significantly more scattered behaviour with larger spacing between the levels despite both using ^{100}Sn as cores. Work is ongoing to extend these calculations to a larger valence space.

With the spectroscopic accuracy achieved with these *ab-initio* calculations it appears that the ground-state level ordering of the nearly degenerate orbitals $2d_{5/2}$, $1g_{7/2}$ remains a significant challenge for these calculation methods. It is however interesting to note that all calculations clearly favour a $7/2$ ground-state for ^{101}Sn .

8.5.2 g-factors

The VS-IMSRG calculations of magnetic dipole moments are presented in terms of g-factors for states having spin $5/2$, $7/2$ and $1/2$, together with the experimental data in Fig. 8.13. Since the ground-state spins have not yet been firmly established for $^{101,103}\text{Sn}$ calculated g-factors assuming both a $7/2$ and $5/2$ ground-states are presented.

Calculations using the EM1.8/2.0 interaction shows a remarkable agreement with experimental $5/2$ g-factors when assuming a ^{100}Sn core, as indicated with orange symbols in the figures. Predictions for the $5/2^+$ states of ^{101}Sn and ^{103}Sn are also similar, as expected. These calculations were also performed using the ^{80}Zr core, indicated in red. The much larger valence space had little influence on the calculated g-factors for $^{101,103}\text{Sn}$, however, a notable deviation was seen for ^{105}Sn . This could be caused by the lack of complete convergence. The indifference to the use of a larger valence space compared to when using a ^{100}Sn indicates that the nucleons within the ^{100}Sn core have little influence on the calculated g-factors.

The calculations using N2LO_{so} with a ^{100}Sn core also show an overall good agreement with experiment, except some minor peculiarities. The g-factors for the $7/2$ states and the $5/2$ state of ^{109}Sn are in even better agreement with the experimental data compared to the calculations using the EM1.8/2.0 interaction. However, an unintuitive g-factor trend is observed for the spin $5/2$ states. The trend diverge to g-factors corresponding to a that expected for a $7/2$ state. Upon further inspection it turns out that the effective operator from the two interactions remain the same although the transition densities are quite different. This indicates that the two interactions provide the significantly different valence-space wave functions. Especially, it was found that the EM 1.8/2.0 can reproduce the nearly degenerate levels of $2d_{5/2}$ and $1g_{7/2}$ orbitals, while the N2LO_{so} cannot. A careful investigation using larger model space and convergence tests are ongoing to investigate this further.

It is important to note that throughout these calculations the bare proton and neutron, orbital and spin g-factors, are used, contrary to the shell-model calculations where single-nucleon g-factors are typically quenched to account for missing interactions in the chosen model space. The remarkably accurate prediction of magnetic moments is astonishing considering the complexity of the nucleus. These results reveal the current progress for predicting nucleon orbital properties of nuclei as heavy as tin.

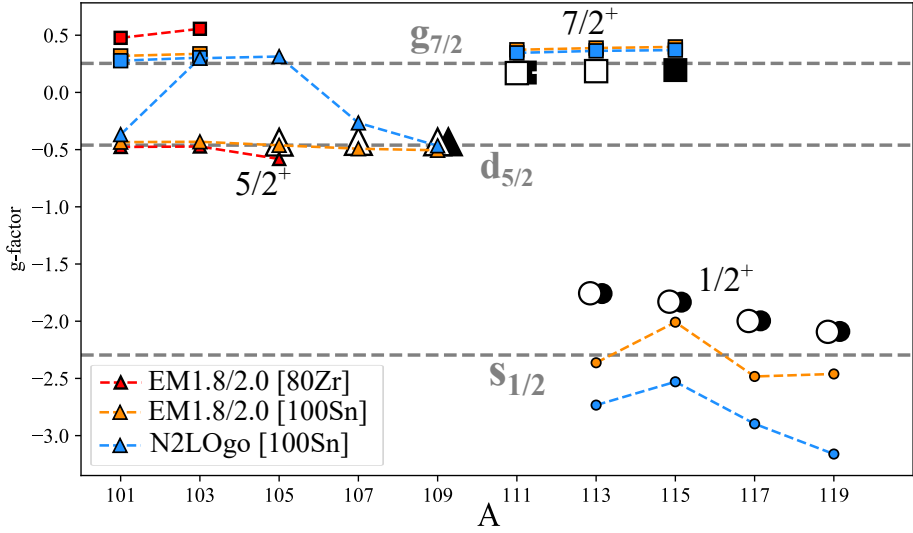


Figure 8.13: Comparison between measured and calculated g-factors using VS-IMSRG with different interactions and valence spaces (see text for more details). The white markers indicate measurements from this work and the black show literature values as indicated earlier.

8.5.3 Quadrupole moments

The evolution of the calculated quadrupole moment for neutron-deficient tin isotopes in the $5/2^+$ or $7/2^+$ states are compared with experiment in Fig. 8.15 and 8.14 respectively.

When considering isotopes in the $7/2^+$ state, the experimental quadrupole moments from $^{115m,113m,111}\text{Sn}$ reveal a reduced quadrupole moment going from $^{115m,113m}\text{Sn}$ to ^{111}Sn . The VS-IMSRG calculations using both interactions are in close agreement with each other for these three isotopes, but underestimate the quadrupole moment by approximately a factor of two compared to experiment. Since the E2 operator used for calculating the quadrupole moment includes a dependence on the nuclear radius, which is typically underestimated in this mass region, the quadrupole moments are often underestimated as well [27]. The calculated values using the N2Logo interaction suddenly jumps at ^{109}Sn . This is likely associated with the sudden orbital flip as seen in Fig. 8.12. Despite this jump both interactions predict almost linear reducing trends towards a negative quadrupole moment for ^{101}Sn where both interactions again converge.

On the other hand, the neutron-deficient isotopes assuming the $5/2^+$ state, shown in Fig. 8.15, reveal a large discrepancy between the two interactions used. The calculations using the EM1.8/2.0 interaction reveal a quadrupole moment 20 times weaker than indicated by experiment for ^{109}Sn , with a 0 crossover point at ^{107}Sn as predicted according to a generalized seniority filling of $1g_{7/2} \otimes 2d_{5/2}$. The underestimated quadrupole moment above ^{101}Sn is not unreasonable considering the small valence space used.

As for the calculated quadrupole moments using the N2LO_{sg} interaction an erratic behaviour is observed which is consistent with the difficulties faced earlier with the orbital ordering. Despite this both interactions are in agreement for ^{101}Sn where the calculation converge with the single particle effective quadrupole moment. The negative quadrupole moment of ^{101}Sn is in agreement within uncertainty of the single particle value predicted by the seniority filling of $2d_{5/2}$, as discussed earlier in Sec. 8.2.

It is likely a larger valence space with an open ^{100}Sn core is necessary to accurately explain the quadrupole moment above ^{100}Sn as the correlations with proton pairs in the $1g_{9/2}$ orbital are expected to have an influence of the quadrupole moment for these isotopes. Calculations using a larger valence space with a ^{80}Zr are currently in progress at the time of writing using both interactions.

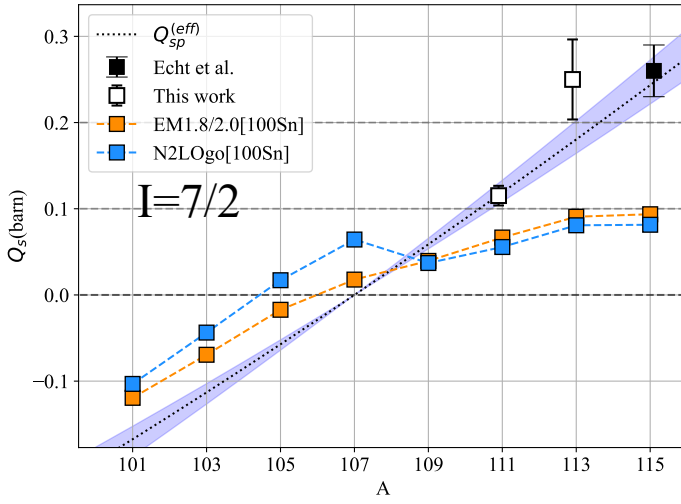


Figure 8.14: Comparison between experimental and calculated electric quadrupole moments for isotopes in the $5/2^+$ state. The single particle quadrupole moment according to the generalized seniority filling of a $1g_{7/2} \otimes 2d_{5/2}$ configuration is presented, as discussed in Sec. 8.2.

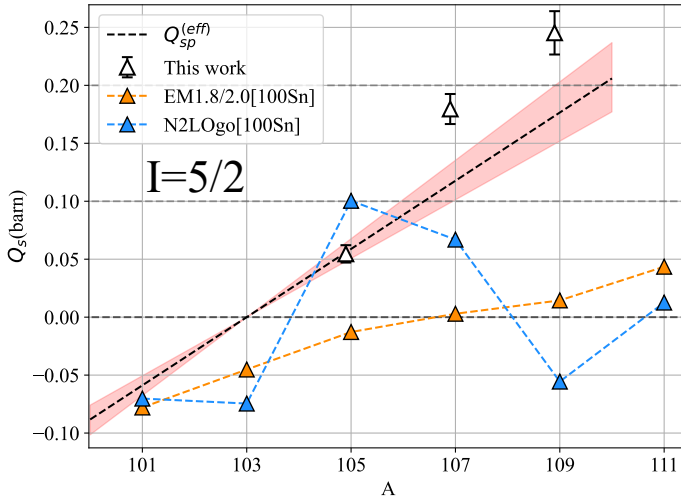


Figure 8.15: Comparison between experimental and calculated electric quadrupole moments for isotopes in the $5/2^+$ state. The single particle quadrupole moment according to the seniority filling of the $2d_{5/2}$ state is presented, as discussed in Sec. 8.2.

8.5.4 Nuclear charge radii

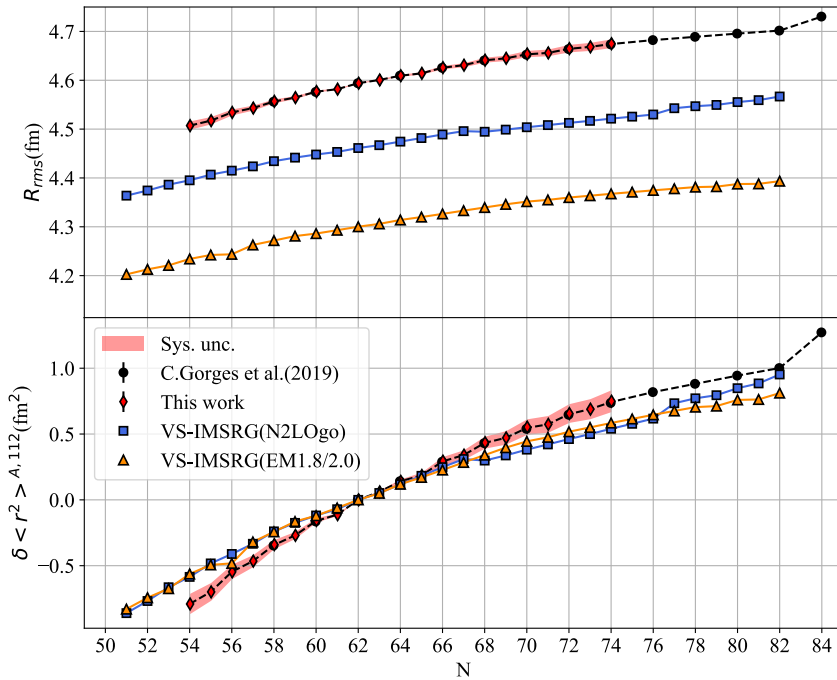


Figure 8.16: Comparison between experimental and calculated rms charge radii of tin isotopes using VS-IMSRG with two ab-initio based interactions. The changes in mean-square charge radii are presented using ^{112}Sn as reference.

In the last decade charge radii of increasingly more complex nuclei have provided particularly stringent tests for bench-marking nuclear theory [141, 130, 164]. The nuclear interactions used in this work have been used successfully to describe different regions of the nuclear chart and is reaching a level where it can compete with phenomenological models. The N2Logo interaction showed particularly good agreement describing the trend for potassium isotopes as presented by Koszorus *et al.* [130], while the EM1.8/2.0 interaction provided a satisfactory description of the microscopic features along the copper isotopic chain as presented by de Groote *et al.* [164].

In order to tackle the new stepping stone in the nuclear landscape both EM1.8/2.0 and the N2Logo interactions were used in VS-IMSRG calculations using a ^{100}Sn core for calculating the charge radii of ^{101}Sn up to ^{132}Sn . The calculated rms charge radii and mean-square charge radii are presented together

with the experimental data in Fig. 8.16. Both interactions slightly underestimate the rms charge radii, with a discrepancy of 0.1 fm for N2Logo and 0.3 fm when using EM1.8/2.0. This is a common trait of these types of calculations due to the difficulties of accurately predicting the nuclear saturation density [164]. While the calculations roughly predict the parabolic trend between shell closures the slope of their trends are slightly underestimated. The calculated trends are up to 3σ off the systematic uncertainties from the atomic factors which indicates that something is likely missing in the calculations. Furthermore, while the overall experimental trend is smooth on the scale of the presented figure there are notable artifacts in these calculations. There is a clear kink at ^{106}Sn when using the EM1.8/2.0 interaction which could originate from the difficulties caused by the issues from the HF level ordering discussed earlier. Another reason could be that the calculations have not yet converged for this isotope. Likewise, N2Logo shows sudden steps at $N = 68$ and $N = 77$ which are unexplained at present.

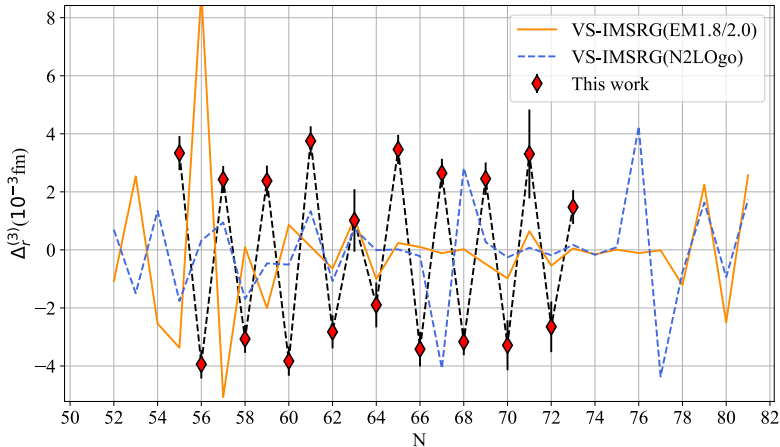


Figure 8.17: A comparison of the calculated odd-even staggering effect in tin compared with experimentally measured radii provided in this work.

The finer details of these trends are revealed by looking at the three-point average (Eqn. 7.1), seen in Fig. 8.17. Neither of the two interactions are able to reproduce the observed odd-even staggering of the measured isotopes. Seemingly both calculations predict almost zero odd-even staggering in between the shell closures (ignoring the jumps seen at $N = 68$ and $N = 77$ for the N2Logo interaction). Interestingly, both calculations seem to converge to a normal odd-even staggering below the neutron heavy shell closure at $N = 82$. On the other side when approaching ^{100}Sn , calculations predict an increasing OES but

with a clear disagreement between both interactions as well as experiment. This could mean that a larger valence space is required where excitations beyond the shell gap are allowed, in order to more accurately reproduce the detail trends in radii approaching ^{100}Sn .

8.5.5 Summary of *ab-initio* calculation results

Both the microscopic and macroscopic nuclear electromagnetic properties of tin isotopes have been used for benchmarking preliminary results from state-of-the-art VS-IMSRG calculations using two different *ab-initio* interactions. Considering that the nuclear interactions employed here are only constrained to experimental data up to $A \leq 4$ and no quenching is used, the accuracy achieved when describing the microscopic g-factors of tin nuclei is remarkable. This shows that these *ab-initio* interactions to a good degree account for the physics required to describe the magnetic moment generated by the unpaired neutron as well as the residual currents generated from the pions and nucleons in the core.

While the nuclear magnetic dipole moments are in overall good agreement, many discrepancies between calculations and experiment were revealed. As it turns out, the nearly degenerate $2d_{5/2}$ and $1g_{7/2}$ orbitals are particularly challenging for these types of calculations due to their small energy separation. Deficiencies in the HF based level ordering have been revealed which limit the spectroscopic precision of these types of calculations which are essential for understanding the ordering of the nearly degenerate orbitals above ^{100}Sn . An investigation is ongoing on how this issue can be resolved based on these results. Currently, all calculations clearly favor a $7/2$ ground-state in ^{101}Sn which is in contradiction to the continued $2d_{5/2}$ ground-state ordering suggested earlier in Sec. 8.1.

The calculations significantly underestimated the quadrupole moment when using a locked ^{100}Sn core. This was expected considering that excitations of protons in the $1g_{9/2}$ orbital have been suggested to have an important contribution in describing the enhanced quadrupole moment for the neutron-deficient tin isotopes. Calculations using a larger model space with a ^{80}Zr core are ongoing at the time of writing. Likewise, the trends in nuclear charge radii are also underestimated. These are remaining challenges which indicate that some physics remain missing in these calculations.

We conclude from these results that the EM1.8/2.0 interaction is in better overall agreement compared to the N2Logo interaction in the tin region. However, the N2Logo interaction is in closer agreement when compared with the absolute charge radii as also seen in other regions of the nuclear chart [51].

Chapter 9

Perspectives for future experiments

While many interesting new insights of the nuclear structure properties of the lightest tin isotopes have been revealed, equally many questions remain which could be answered from spectroscopic measurements of isotopes below ^{104}Sn . It is concluded from the discussion in Sec. 8.3 that measurements of the hyperfine structures of $^{101,103}\text{Sn}$ will be able to finally settle the controversy regarding the ordering of the $2d_{5/2}$, $1g_{7/2}$ neutron orbitals and could directly confirm or deny the suggested shell-model filling of the $2d_{5/2}$ orbital above ^{100}Sn . Furthermore, the continued measurements of charge radii would reveal if the rapidly reducing charge radius with removed neutrons continues as predicted towards ^{100}Sn with further insight into the continued trend of the odd-even staggering.

In this chapter the main challenges which prevented the study of more neutron-deficient isotopes, are described. The attempt at resolving the hyperfine structure of ^{103}Sn is used as an example to reveal the limitations faced during the experiment. Suggested improvements for enhancing the ionization efficiency and reducing the background contamination are presented together with a feasibility study of future spectroscopic measurements reaching down to ^{101}Sn using the CRIS technique.

9.1 Attempted spectroscopy of ^{103}Sn

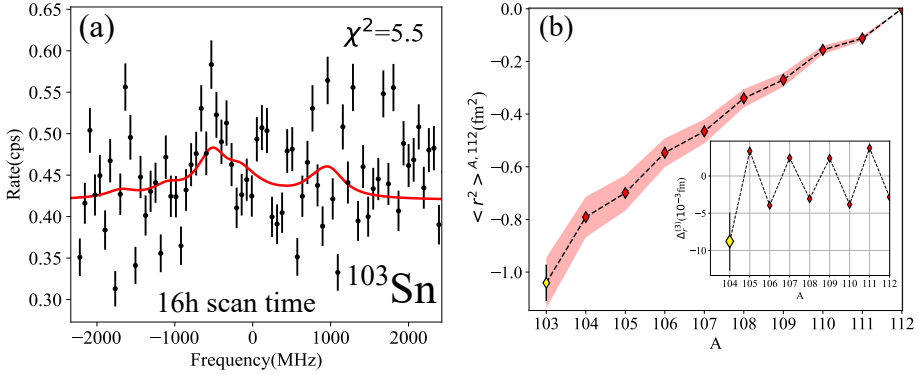


Figure 9.1: (a) show a 16 h CRIS measurement of ^{103}Sn (65 MHz binsize) using the PP transition scheme together with the fitted function in red. The mean-square charge radii including the value for ^{103}Sn is presented in figure (b).

Table 9.1: Measured isotope shift of ^{103}Sn using the PP transition with calculated mean-square charge radii and rms charge radii using FS and MS factors presented in Tab. 7.8.

Isotope	$\delta\nu^{103,112}$ (MHz)	$\langle r^2 \rangle$ (fm ²)	R_{rms} (fm)
103	-2247(138)	-1.041(69)[96]	4.479(8)[11]

A significant time during the online experiment was dedicated to the attempt at resolving the hyperfine structure of ^{103}Sn , see Fig. 7.1. The combined data from a 16h measurement using the PP transition with repeated scans across the predicted scan region, are presented in Fig. 9.1(a). A hint of a resonance peak was revealed when merging all data which remained despite varying bin size. This structure was fitted by constraining the hyperfine parameters which were calculated assuming $\mu_I = 1.081$, $I = 5/2$ and $Q_s = 0$ for $^{103}\text{Sn}^1$. Furthermore, both the FWHM² and Racah intensities were fixed. The model is seen fitted to the spectrum in Fig. 9.1(a) which provided the IS relative to ^{112}Sn and thereafter its mean-square charge radius, presented in Tab. 9.1. In Fig. 9.1(b)

¹These predictions were based on the assumed continued single-particle trends of nuclear moments described in Sec. 8.1.

²A linewidth of 450 MHz was measured for ^{112}Sn under identical conditions which was used as constraint in the fit of ^{103}Sn .

the rms charge radius of ^{103}Sn is added to the trend in charge radii along the tin chain. A continued downward trend following the normal odd-even staggering is revealed as highlighted in the inset figure. However, the large statistical uncertainty and unresolved structure makes it difficult to draw any conclusion based on this observation. Nevertheless, the tempting hint provides a strong motivator for future spectroscopic measurements to verify this observation.

Assuming identical conditions to the experimental conditions in this work, how much time would have been required to resolve the structure of ^{103}Sn ? In order to answer this question the spectrum of ^{103}Sn was simulated with the experimental conditions observed in 9.1(a) for different scan times of 16h, 4 days and 12 days, see Fig. 9.2. Despite 12 days of continuous scanning the hyperfine structure would only be partially resolved. The measurement is clearly swamped by the overwhelming background which correspond to roughly 80% of the total signal rate. Measurements were performed with the HRS magnet set for ^{102}Sn to investigate the background rates here. A significant background rate of 0.7(2) cps was measured at CRIS. Since the yields are expected to be roughly ten times lower than ^{103}Sn it was concluded that despite lacking a hyperfine structure this measurement was unfeasible under the current conditions. It is thus suggested that the background signal needs to be reduced in order to realize spectroscopic measurements of the more exotic neutron-deficient tin isotopes.

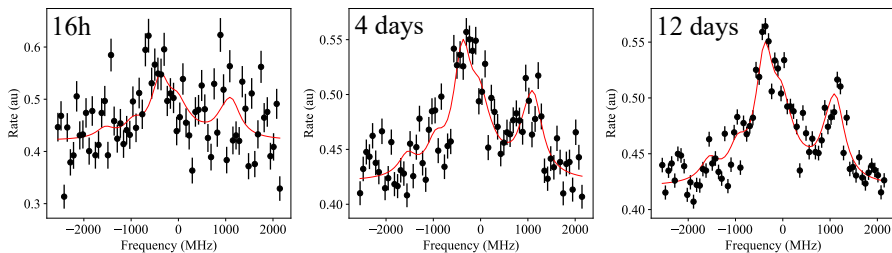


Figure 9.2: Simulated spectrum of ^{103}Sn at different scan duration assuming similar conditions to the experiment presented in this work.

9.2 Background due to contamination

Isobaric contamination makes up an increasing percentage of the beam content towards the more exotic neutron-deficient isotopes, as seen in Fig. 7.3. Over 80% of the beam content was composed of surface ionized contamination in ^{105}Sn , this fractional contamination increases significantly for the more exotic isotopes. A large fractional beam contamination increases the collisional

ionization occurring after the deflector plates which are detected in the ion detector as background events. This contamination was expected to be largely composed of indium isobars due to the low IP compared to the neighbouring elements, as shown in Fig. 4.5.

When searching for ^{105}Sn resonances using the two-step SP transition scheme, a series of strong resonances were observed as seen in Fig. 9.3. It was concluded that the observed resonances originated from surface ionized contamination as they remained despite blocking RILIS lasers. These resonances likely originated from the large hyperfine structure of ^{105}In ($I=9/2$) isobar which reached the scanning range of the transition studied in ^{105}Sn . In indium there are mainly two fine structure states $5s^2 5p^2 P_{1/2,3/2}$ populated after the CE process [148]. Electrons in these two populated states could have been excited with the first laser step to higher-lying states expected at either $35544.0(5) \text{ cm}^{-1}$ or $37756.5(5) \text{ cm}^{-1}$. However, only the latter could have been non-resonantly ionized using the 1064 nm light. This predicted level at $37756.5(5) \text{ cm}^{-1}$ in indium could not be found in either the NIST [147] or the Kurucz [216] databases. Spectroscopic measurements are therefore required to verify if this state exists in indium. Accurate determination of the contamination within the beam can be performed by separating different masses with a resolving power of better than 10^5 using Multi-Reflection Time-of-flight mass spectroscopy (MR-TOF MS) measurements of the beam, regularly performed at ISOLTRAP experiment at ISOLDE [217]. MR-TOF MS measurements were performed on ISOLDE beams of neutron-deficient indium isotopes produced from a similar LaC_x target used for the production of tin isotopes presented in this work. These measurements indicated a significant contamination caused by SrF^+ molecular ions [218] which likely also contribute to a large fraction of the surface ionized background contamination within the beam seen in this work. This section reveals that it is more than just reducing collisionally ionized background events, laser ionization of other species can also result in difficulties during a CRIS measurement.

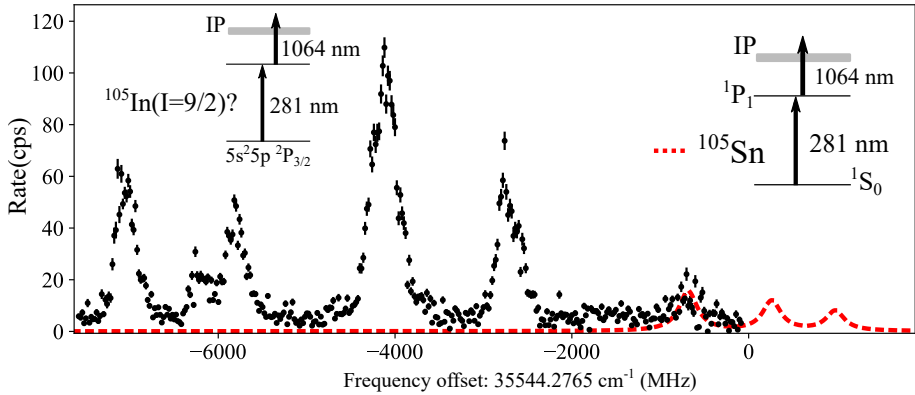


Figure 9.3: While searching for ^{105}Sn resonances using the SP transition resonances of surface ionized contamination were observed. These resonances were likely originating from a hyperfine structure of ^{105}In .

9.3 Suggested approaches for future measurements

In order to measure the more exotic neutron-deficient tin isotopes using the CRIS technique both the efficiency and background suppression need improving. As described in Sec. 4.9 the efficiency achieved at CRIS is highly element specific due to the the neutralization efficiency and the population in the probed atomic state. A relatively small fraction of only 20% was neutralized in the CE process, this together with low relative population in the probed atomic state (17% in the 3P_2 state as seen in Fig. 4.10) results in a loss of almost 97% of the tin beam.

The CE simulations presented in Fig. 4.10 indicate a significantly greater final population in the 3P_1 state of 29% which could enhance the efficiency by almost a factor of 2 compared with the probed 3P_2 state. However, simulations performed before the online experiment estimated an almost identical population in the 3P_2 and 3P_1 states when using potassium neutralizer. The difference between the old and new CE calculations is expected to originate from slight differences in the electron capture cross sections and transition rates used in the calculations. When using potassium neutralizer a considerable initial population are expected in the short-lived (ns lifetime) states which quickly decay into the lower lying states. The final population is therefore particularly sensitive to the transition rates and decay channels in the calculation. Experiments therefore need to confirm if the efficiency can indeed be enhanced by probing from the 3P_1 state compared to the 3P_2 state. The population of an atomic level can be

enhanced with optical pumping from other populated states, as presented in the work by Hanna Berg [219]. The population in the 1S_0 using sodium neutralizer can be enhanced by over 50% by optical pumping of the $5p^2\ ^1D_2 \rightarrow 5p5d^3P_1$ transition followed by a decay into the 1S_0 state.

Three suggested approaches are presented below which could improve the efficiency and background suppression for the spectroscopic study of tin beams at CRIS:

- Ion-ionization spectroscopy:** One way to remove the losses associated with the neutralization process is to remove the need for a CEC altogether, and perform spectroscopy directly from the ground-state of the ions entering CRIS [140]. Multiple resonant laser steps could be used to perform ion-ionization from the ionic ground state in Sn^{1+} resulting in doubly-ionized tin Sn^{2+} , using a suggested four-step resonance ion-ionization scheme is shown in Fig. 9.4(a). The resonantly 'double'-ionized 2^+ ions could then be selected after the bend and detected in the ion detector. This procedure would in principle eliminate all losses associated with the neutralization process. This would also remove vapour contamination within the interaction region originating from the CEC, which further reduces collisionally ionized background events. Feasibility studies were performed on beams of Ba^3 which indicated that the selectivity in separating the 2^+ from 1^+ ions at CRIS was insufficient in the current configuration. Furthermore, the lasers required to perform the resonance ion-ionization of tin were not available at CRIS during this work. Future upgrades are planned which will improve the ion selectivity.
- Negative ion neutralization:** Tin is known to have an extraordinarily high negative ion conversion efficiency from CE interaction [220, 221]. Measurements of 50 keV Sn^{1+} beams passing CEC containing Cs vapour resulted in an over 40% negative ion conversion efficiency [221]. It is likely that a significant fraction of the tin beam passing the CEC is composed of Sn^{1-} resulting from a two-step CE interaction with the potassium vapour. Any generated negatively charged ions were deflected after the CEC and 'lost' during the online measurements. As discussed in M. Vandevraye et al. [222] a single laser can be used to photo detach the electron and resonantly populate the selected $^3P_{0,1,2}$ states in the neutral Sn as seen in the level scheme in Fig. 9.4(b). This laser could neutralize the negative ions before the interaction region. This would result in an enhanced neutralization efficiency while also selectively populating the probed state in the atom. Assuming a 40% negative ion conversion efficiency after the

³Barium was chosen since it has the lowest ion-ionization threshold of all stable elements at 10 eV, reducing the number of required resonant laser steps.

CEC this would result in an over 12x enhanced detection efficiency when probing from the 3P_2 state in the atom.

- **Field-ionization from Rydberg states:** During this thesis work a field-ionization unit was installed after the interaction region and successfully used to resonantly ionize indium atoms from selectively populated Rydberg states, described in detail in Ref. [223]. This device is comprised of closely spaced metallic grids placed in the beam path at the end of the CRIS interaction region. By applying a high-voltage potential across the grids creates a localized region with a high-electric field gradient, sufficient to ionize atoms populated in Rydberg states, as described in section 4.4. Any collisionally ionized events occurring before the field-ionization unit can therefore be removed using an additional set of deflection electrodes. This field-ionization unit also removes the need for a high-powered non-resonant laser step which significantly reduces non-resonantly photo-ionized background events. Suggested laser steps for selectively populating Rydberg states in tin is presented in Fig. 9.4(c). This device is in the process of being installed at CRIS at the time of writing.

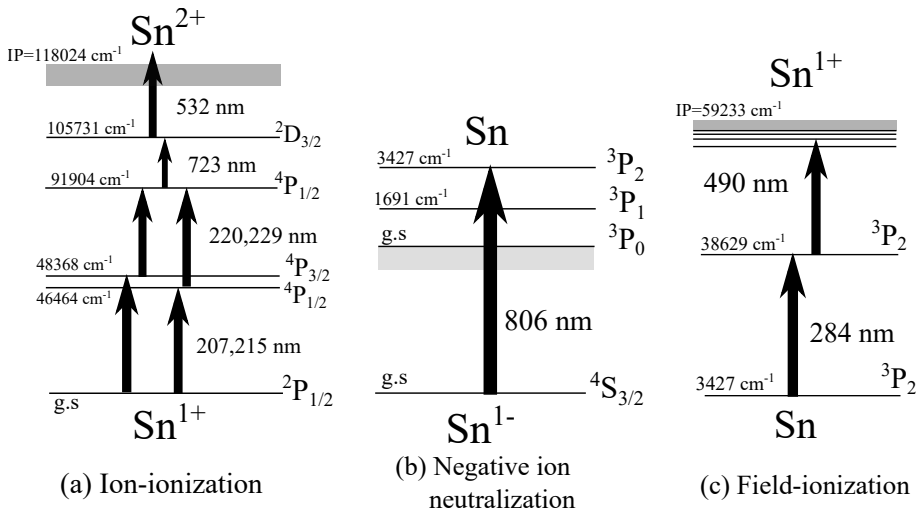


Figure 9.4: Suggested resonant ionization schemes of tin.

The enhanced efficiency resulting from negative ion neutralization could be merged together with the background suppression by field-ionizing from selected Rydberg states. An efficiency above 1% could potentially be achieved with an estimated background suppression of at least an order of magnitude. However,

measurements are required in order to verify the negative ion production after the CEC and to confirm the background suppression achieved with the installation of the field-ionization unit at CRIS.

Additionally, a radio frequency cooler buncher (RFQCB) is in currently under development and planned for installation before the CRIS beamline, described in [224]. This dedicated RFQCB will further increase the data collection efficiency while simplifying the process of obtaining reference mass measurements during an experiment. There are also plans of constructing a compact MR-TOF MS which would enable direct mass identification and filtration of the beam content which would eliminate any unwanted contamination from the beam before spectroscopic measurements are performed at CRIS.

9.4 Simulated spectrum of $^{101,102,103}\text{Sn}$

Fig. 9.5 shows the simulated hyperfine spectra of $^{101,102,103}\text{Sn}$ probing the $5p^2\ ^3P_2 \rightarrow 5p6s\ ^3P_2$ transition. Here an efficiency of 1% is assumed with 10x background suppression for ^{103}Sn and ^{102}Sn and a 100x background suppression for ^{101}Sn relative to the background conditions observed from measurements of ^{103}Sn and ^{102}Sn , presented earlier in Sec. 9.1. The hyperfine structures for the $^{103,101}\text{Sn}$ are simulated assuming both nuclear spin 5/2 and 7/2 with nuclear moments suggested from continued trends presented in Sec. 8.5⁴. The simulated spectra reveal that with the suggested improvements presented earlier in Sec. 9.3 measurements of ^{103}Sn and ^{102}Sn are feasible during an experimental campaign lasting roughly 1 week. However, significant time is required in order to resolve the hyperfine structure of ^{101}Sn where over 7 days of continuous scanning is barely enough to resolve the structure despite a 100x suppression in background.

⁴These simulations done for ^{103}Sn with ($I = 5/2$, $\mu = -1.081$, $Q_s = 0$) and ($I = 7/2$, $\mu = 0.6$, $Q_s = -0.05$) and for ^{101}Sn using ($I = 5/2$, $\mu = -1.081$, $Q_s = -0.05$) and ($I = 7/2$, $\mu = 0.6$, $Q_s = -0.06$).

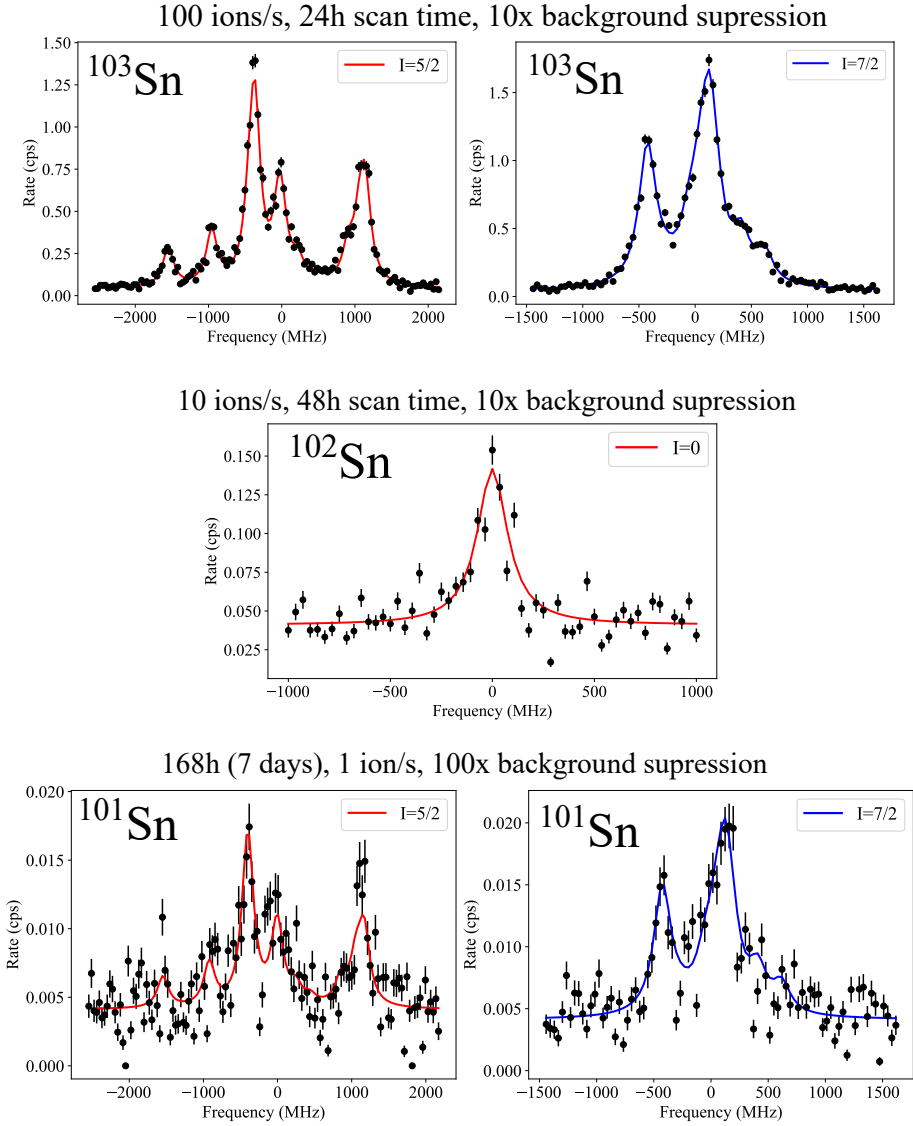


Figure 9.5: Simulated hyperfine structures of $^{103,102,101}\text{Sn}$ probing the $5p^2\ ^3P_2 \rightarrow 5p6s\ ^3P_2$ transition, assuming 1% efficiency with 10x and 100x suppressed background signal.

9.5 Summary

The main limiting factor which prevented measurements of more exotic tin isotopes below ^{104}Sn resulted from the surface ionized background contamination, which makes up an increasing fraction of the beam content for the more exotic isotopes (suggested to be dominated by isobaric indium and SrF contamination). The efficiency of future measurements could potentially be improved by a factor of 12 by resonant photo-detachment of electrons from negatively charged tin ions produced in the CE-process. In addition, by performing field-ionization from Rydberg states in tin using a field-ionization unit could suppress non-resonantly ionized background events. With these improvements measurements of ^{103}Sn and ^{102}Sn would likely be feasible. A significant effort is required in order to measure ^{101}Sn and ^{100}Sn with very low production yields expected below <1 ion/s.

A considerable work is ongoing on the target side to improve the selectivity such as Laser Ion Source and Trap (LIST) and ion source which could suppress the isobaric contamination by over a factor of 10^6 at the expense of a loss in efficiency, presented in the work by D.A. Fink *et al.* [225]. Furthermore, long term goal at CERN will allow 2 GeV protons with an increased beam current. ABRABLA simulations presented by Ramos *et al.* [134] reveal a potential improvement of a factor of 10x gain of neutron-deficient such as ^{100}Sn using a liquid La target. With these additional improvements laser spectroscopic measurements of ^{101}Sn and ^{100}Sn could become possible in the near future at ISOLDE.

Chapter 10

Summary and conclusions

In this work nuclear electromagnetic properties (spins, magnetic dipole moment, electric quadrupole moment and charge radii) of neutron-deficient tin isotopes have been established down to ^{104}Sn thanks to the experimental development of the CRIS technique together with advancements in quantum chemistry calculations.

The new understanding of the probed electronic configuration in tin from both theory and experiment provided the tools necessary to extract the nuclear moments and charge radii from the acquired data. These results allowed the first nuclear model-independent spin assignment of ^{107}Sn and ^{105}Sn . The measured nuclear moments of ^{105}Sn is in good agreement with a single neutron hole configuration in the $2d_{5/2}$ neutron orbital, expected for a sequential shell-model filling of a $2d_{5/2}$ neutron orbital above ^{100}Sn assuming a strong shell closure.

The unpaired neutron remains in the $2d_{5/2}$ neutron orbital while neutron pairs are added in the $1g_{7/2}$ orbital up to ^{109}Sn . Such configuration is favored due to the enhanced pairing energy in the $1g_{7/2}$ orbital as compared to the $2d_{5/2}$ orbital. The enhanced quadrupole moment observed with added neutron pairs in the $1g_{7/2}$ orbital is explained by the proton neutron correlation when allowing protons to scatter across $Z = 50$ from the $1g_{9/2}$ orbital. This enhanced collectivity, observed earlier in through the $B(E2)$ of ^{106}Sn , ^{108}Sn and ^{110}Sn is confirmed here in the spectroscopic quadrupole moments of $^{107,109}\text{Sn}$.

Measurements of nuclear charge radii reveal that the expected parabolic trend between $N = 50, 82$ shell closures is in fact broken at $N = 64$ by a subshell closure which separates the $2d_{5/2}, 1g_{7/2}$ from the $3s_{1/2}, 2d_{3/2}, 1h_{11/2}$ neutron orbitals. A bump in radii observed below $N = 64$ ends in a steep decline

down to ^{104}Sn , with a significantly smaller radius than expected according to a continued parabolic trend between $N = 50, 82$. The bump in radii around masses $^{106-110}\text{Sn}$ is in good agreement with the enhanced collectivity observed in the $B(E2)$ and the enhanced quadrupole moment from the added neutrons in the $1g_{7/2}$ orbital. The steep decline in radius towards ^{104}Sn suggests reduced collective effects towards ^{100}Sn , which points to an increased rigidity of ^{100}Sn .

The new understanding of the electromagnetic properties of tin isotopes provide particularly challenging benchmarks for state-of-the-art ab-initio calculations. The VS-IMSRG framework was used together with two interactions founded on χEFT for interpreting the electromagnetic moments of the tin isotopes. While these calculations struggle with predicting the nearly degenerate level ordering of the shell-model orbitals, the calculated g-factors are in remarkable agreement considering these interactions are only fitted to electromagnetic properties of light nuclei $A \leq 4$ and that no quenching is required. This indicates that the essential single particle orbital motion and the residual interactions are well described using these χEFT based interactions. However, the macroscopic collective properties such as the rms charge radii and electric quadrupole moments are underestimated using both interactions assuming ^{100}Sn core. While these calculations are preliminary the results in the neutron-deficient tin region turns out to be particularly challenging, partly owing to the nearly degenerate level ordering of the lowest lying shell model orbitals.

In order to confirm or disprove the proposed shell-model filling of the $1d_{5/2}$ orbital above ^{100}Sn laser spectroscopic measurements of $^{103,101}\text{Sn}$ are of particular interest. In the outlook a tempting hint of ^{103}Sn resonances were revealed. However, background contamination hindered the measurements of the more neutron-deficient isotopes. Based on the observed limitations improvements are suggested which could make measurements of the more exotic isotopes feasible in the near future.

In conclusion, the electromagnetic properties of the tin atom and in its nucleus have been investigated experimentally and used as a testing ground for state-of-the-art description of both atomic and nuclear structure properties. These challenging benchmarks provided in this work contributes to a small step in the direction towards a unified description of the atom and its nucleus.

Appendix A

Appendix

A.1 Stable tin isotopes

Table A.1: Stable tin isotopes available from the laser ablation ion source using a natural tin target.

Isotope	Abundance (%)	I^π
¹¹² Sn	0.97	0
¹¹⁴ Sn	0.66	0
¹¹⁵ Sn	0.34	1/2 ⁺
¹¹⁶ Sn	14.54	0
¹¹⁷ Sn	7.68	1/2 ⁺
¹¹⁸ Sn	24.22	0
¹¹⁹ Sn	8.59	1/2 ⁺
¹²⁰ Sn	32.58	0
¹²² Sn	4.63	0
¹²⁴ Sn	5.79	0

A.2 Mass identification from TOF measurements at CRIS

Accurate measurements of the ion arrival time in the MagneTOF detectors allow TOF measurements at CRIS. Since the beam energy is defined from the applied acceleration voltage, the TOF information acquired with every measurement can be used for estimating the mass of the detected ion. The measured TOF ($t_{measured}$) of an ion with mass m can be estimated with the following expression:

$$t_{measured} = t_{Ext.} + \frac{L}{\sqrt{2eU}}\sqrt{m}. \quad (\text{A.1})$$

Where $t_{Ext.}$ is the time from the start trigger until the ion reaches its final velocity, U is the ion source acceleration voltage and L is the distance from where the ion reaches its final velocity, to the ion detector. While the beam travel distance can be determined accurately the extraction time $t_{Ext.}$ is unknown. This term includes the mass dependant acceleration time and can vary depending on the extraction voltage and the ablation process.

Two MagneToF ion detectors are installed in the CRIS beamline, the main MagneToF ion detector 2 is located roughly 7 m from the ion source and another MagneTOF detector is located in the differential pumping region, 2.35 m upstream from detector 1, see Fig. 4.3. By performing measurements of the ion TOF at these two locations a more accurate TOF $\Delta t_{1 \rightarrow 2}$ can be determined which is not dependant on the extraction time $t_{Ext.}$,

$$\Delta t_{1 \rightarrow 2} = \frac{\Delta L_{1 \rightarrow 2}}{\sqrt{2eU}}\sqrt{m} \quad (\text{A.2})$$

Measurements of $\Delta t_{1 \rightarrow 2}$ is particularly useful for characterizing beam composition when using compound targets in the laser ablation ion source. TOF spectra of a copper ion beam accelerated to 25 keV into the two detectors are displayed together with the calculated TOF using Eqn. A.2 in Fig. A.1. In detector 1 the prompt ion release is observed with a narrow TOF distribution together with two separated TOF peaks corresponding to the thermal release of $^{63,65}\text{Cu}$ separated in TOF. A wide distribution of TOF is observed in detector 2 where the two isotopes can no longer be separated.

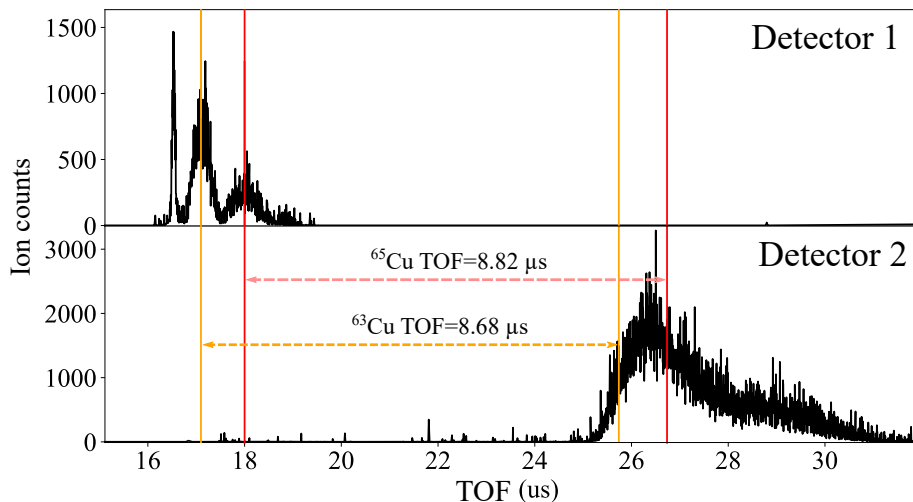


Figure A.1: TOF spectrum for a Copper beam produced from the laser ablation of metallic Copper target accelerated to 25 keV beam energy. The TOF between detector 1 and 2 is calculated using Eqn. A.2 based on the TOF of the observed peaks in detector 1.

Compound ceramic targets have also been used in the CRIS ion source. A BaF^2 compressed pellet target was created in order to facilitate measurements of molecular beams at CRIS. The TOF spectrum using a BaF^2 ceramic target at 20 keV beam energy is presented with the calculated TOF of Ba, BaF and BaF^2 for identifying the composition of the compound beam, see Fig. A.2.

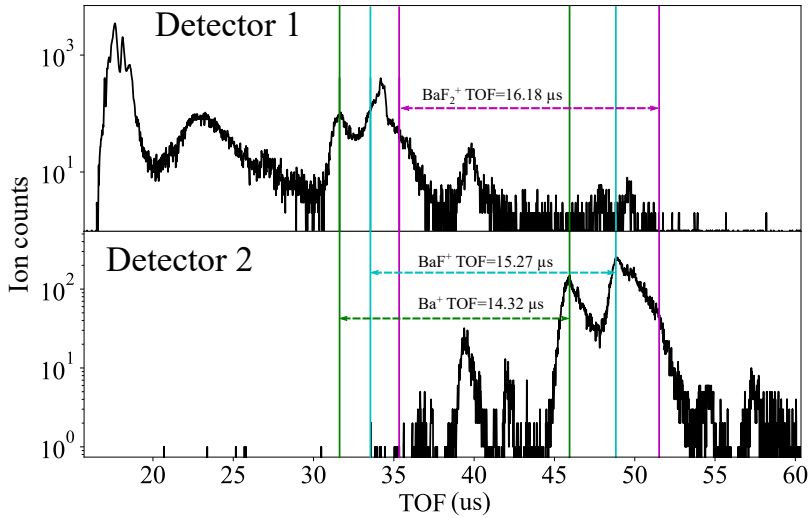


Figure A.2: TOF spectrum for a BaF_2^+ produced from the laser ablation of compressed pellet of BaF_2 and accelerated to 20 keV beam energy. The TOF of Ba and its molecular species are calculated and the suggested peak locations are presented in the figure. The large number of events arriving before 30 us are likely composed of the 2^+ charge states which does not reach detector 2.

A.3 Comparing EFGs using $Q_s(^{109}\text{Sn})$

The determination of spectroscopic quadruple moments require calculated EFGs for the atomic levels. Therefore, the extracted Q_s is strongly dependant on the accuracy of the calculated EFGs. The B_{hf} parameter of ^{109}Sn has been probed in several different atomic states, in this work and other works [77, 76]. The consistent set of EFGs computed using the relativistic FSCC approach presented in paper I were used to calculate Q_s of ^{109}Sn from the experimentally measured B_{hf} values, as presented in Tab. A.2. Here, the upper and lower B_{hf} values of the fitted spectrum using the PP transition were unconstrained by their ratio.

The calculated Q_s from the measured B_{hf} in each transition are all in good agreement within the estimated uncertainty, as displayed in Fig. A.3. This further verifies the reliability of the atomic EFG calculations. The weighted-mean value is in perfect agreement with the constrained value presented in this work in Tab. 7.5.

Table A.2: The table presents the calculated spectroscopic quadrupole moment for ^{109}Sn from experimental B_{hf} parameters together with calculated EFGs. The B_{hf} indicated in bold are taken from [77].

Level	B_{hf} [MHz]	EFG[MHz/b]	EFG Ref.	Q_s [b]
$5p7s^1P_1$	113(11)	378(68)		0.299(29)[54]
$5p^2\ ^3P_2$	-158(20)	-691(28)	[107]	0.229(28)[9]
$5p6s^3P_2$	245(8)	990(40)		0.247(8)[10]
$5p6s^1P_1$	154(5)	645(58)		0.239(8)[21]
		Weighted mean:		0.244(5)[9]
$5p6s^1P_1$	154(5)	703(50)	[226]	0.219(7)[16]
		593(89)	[76]	0.26(8)[39]

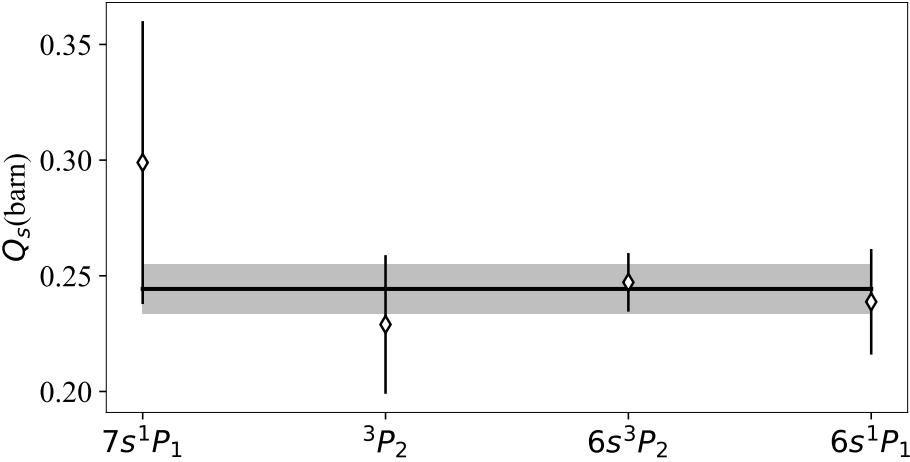


Figure A.3: Quadrupole moment of ^{109}Sn extracted from different atomic states using EFGs from [107]. Solid line indicate the weighted average with uncertainty with grey shade.

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