

Laser spectroscopy of neutron-deficient mercury isotopes and commissioning of a gas-jet based RFQ ion guide

CERN-THESIS-2018-289
// 2018


Simon Sels

Supervisor:
Prof. dr. P. Van Duppen

Dissertation presented in partial
fulfillment of the requirements for the
degree of Doctor of Science (PhD):
Physics

August 2018

Laser spectroscopy of neutron-deficient mercury isotopes and commissioning of a gas-jet based RFQ ion guide

Simon SELS

Examination committee:
Prof. dr. E. Janssens, chair
Prof. dr. P. Van Duppen, supervisor
Prof. dr. M. Huyse
Prof. dr. G. Neyens
Prof. dr. R. Raabe
Dr. R. Ferrer

Dissertation presented in partial
fulfillment of the requirements for
the degree of Doctor of Science
(PhD): Physics

Prof. dr. A. Andreyev
(University of York, UK.)

August 2018

© 2018 KU Leuven – Faculty of Science

Uitgegeven in eigen beheer, Simon Sels, Celestijnenlaan 200D box 2418, B-3001 Leuven (Belgium)

Alle rechten voorbehouden. Niets uit deze uitgave mag worden vermenigvuldigd en/of openbaar gemaakt worden door middel van druk, fotokopie, microfilm, elektronisch of op welke andere wijze ook zonder voorafgaande schriftelijke toestemming van de uitgever.

All rights reserved. No part of the publication may be reproduced in any form by print, photoprint, microfilm, electronic or any other means without written permission from the publisher.

Dankwoord

Dit is het laatste deel van mijn doctoraat dat ik schrijf en vreemd genoeg ook één van de moeilijkste. Er komt nu een tastbaar einde aan een aangenaam hoofdstuk in mijn leven. Het voelt dan ook aan als vanzelfsprekend een heleboel mensen te bedanken die me gedurende deze periode hebben bijgestaan.

Als eerste en in het bijzonder, wil ik mijn promotor Piet Van Duppen bedanken om mij de mogelijkheid te hebben gegeven een doctoraat te volbrengen in de kernspectroscopiegroep. De vele discussies met Piet, Mark, Rafael, Yuri, Andrei, Anatoly en Taka hebben me enorm veel bijgebracht en ik ben hen heel dankbaar voor de aangename begeleiding die ze me hebben gegeven.

Het IKS was steeds een aangename werkplek. De vriendelijke sfeer die er heerste en de ambitieuze, internationale omgeving waren inspirerend. Ik wil graag de collega's bedanken die mij er thuis hebben doen voelen bij mijn aankomst: Lars, Celine, Nele, Jytte en Jan. Ook Camilo, Dani, Yisel en Ruben, die het met mij tot het einde hebben moeten uithouden, verdienen hier een vermelding voor. Thank you to all the great office mates I had over the last years. I enjoyed all the fun moments we shared, whether it was just having a quick coffee or hearing about your last romantic (Tinder) conquest.

Bedankt André, Matthias, Kristof, Simon, Lars en Dani om me fysiek te laten afzien tijdens onze sportieve middagen en hierbij mee voor te zorgen dat ik het IKS een aantal kilo's lichter verlaat dan hoe ik er startte.

Bedankt ook aan Paul, Hilde, Willy, Bert, Luk, Fabienne, Danielle, Isabelle en het personeel van de technische werkplaats voor hun ondersteuning en hulp.

James and Liam, I am not sure how your names ended up here. If anything, your presence was detrimental for this thesis by having too much fun. All joking asside, thank you guys a lot!

Natuurlijk wil ik ook mijn fantastische familie bedanken.

Aan mijn ouders: bedankt voor alle kansen die jullie me hebben gegeven.

Aan mijn broers, Michiel, Evert en Mathijs: Bedankt voor de prachtige jaren en de hechte band.

En ten slotte: Lieselot, bedankt voor je liefde en steun in alles wat ik doe. Bedankt voor de grote en kleine dingen. Bedankt voor wat je elke dag voor mij betekent.

Simon

Preface

This PhD thesis consists of two parts. Part I describes the results of an experimental campaign studying mercury isotopes at ISOLDE, CERN. Using in-source laser spectroscopy, ground and isomeric-state properties of $^{177-185,202,203,206-208}\text{Hg}$ such as charge radii and electromagnetic moments were measured. The first of two aims of this experiment, was to delineate the region of ground-state shape staggering in the neutron-deficient isotopes around $N = 104$. The second goal was to characterize the charge-radii trend across the $N = 126$ shell closure in the neutron-rich isotopes.

Part II of this thesis describes the design, simulation and commissioning tests of an ion-beam cooler for IGLIS developments at KU Leuven. It has been installed and is operational at the dedicated KU Leuven laboratory for IGLIS studies.

Part 0 of this thesis links the two seemingly unrelated topics of this thesis. Chapter 1 gives an introduction and motivation for the laser-spectroscopy studies on mercury. In chapter 2, Nuclear models that are relevant for the interpretation of the data are introduced briefly. Chapter 3 deals with the theoretical basis and extraction of information from laser-spectroscopy data. In chapter 4, the experimental setup and data analysis are summarized. This chapter also includes two articles that have been written and in which the experimental parts are based on the work described in this thesis. The first publication is accepted and will be published in Nature Physics as

B. A. Marsh, T. D. Goodacre⁺, **S. Sels**⁺, Y. Tsunoda, B. Andel, A. N. Andreyev, N. A. Althubiti, D. Atanasov, A. E. Barzakh, J. Billowes, K. Blaum, T. E. Cocolios, J. G. Cubiss, J. Dobaczewski, G. J. Farooq-Smith, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, L. P. Gaffney, L. Ghys, M. Huyse, S. Kreim, D. Lunney, K. M. Lynch, V. Manea, Y. M. Palenzuela, P. L. Molkanov, T. Otsuka, A. Pastore, M. Rosenbusch, R. E. Rossel, S. Rothe, L. Schweikhard, M. D. Seliverstov, P. Spagnoletti, C. V. Beveren, P. V. Duppen, M. Veinhard, E. Verstraelen, A. Welker, K. Wendt, F. Wienholtz, R. N. Wolf, A. Zadornaya,

and K. Zuber, “Characterization of the shape-staggering effect in mercury nuclei,” *Accepted in Nature Physics*, 2018, ⁺ these authors contributed equally to this work

while the second article is submitted to *Physical Review C* as

S. Sels, T. D. Goodacre, B. A. Marsh, Y. Tsunoda, B. Andel, A. N. Andreyev, N. A. Althubiti, D. Atanasov, A. E. Barzakh, J. Billowes, K. Blaum, T. E. Cocolios, J. G. Cubiss, J. Dobaczewski, G. J. Farooq-Smith, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, L. P. Gaffney, L. Ghys, M. Huyse, S. Kreim, D. Lunney, K. M. Lynch, V. Manea, Y. M. Palenzuela, P. L. Molkanov, T. Otsuka, A. Pastore, M. Rosenbusch, R. E. Rossel, S. Rothe, L. Schweikhard, M. D. Seliverstov, P. Spagnoletti, C. V. Beveren, P. V. Duppen, M. Veinhard, E. Verstraelen, A. Welker, K. Wendt, F. Wienholtz, R. N. Wolf, A. Zadvornaya, and K. Zuber, “Shape staggering of mid-shell mercury isotopes from in-source laser spectroscopy compared with density functional theory and monte carlo shell model calculations,” *To be submitted to Physical Review C*, 2018

The second part of this thesis starts with chapter 5, where the basics of IGLIS are introduced. Chapter 6 describes the principles of an ion-beam cooler. Simulations and design of this RFQ ion guide are detailed in chapter 7. In chapter 8, commissioning and characterizations of the devices operation are tested and compared to results from simulations. The thesis ends with chapter 9, where conclusions of both parts of this work are summarized and outlook to further work is given.

Abstract

The neutron-deficient mercury isotopes are situated in the region of the $Z = 82$ proton shell closure, which is known for its manifestation of shape coexistence. Shape coexistence is the phenomenon in which different degrees or types of nuclear deformation are present at low excitation energy. The mercury isotopes are in fact one of the prime examples of shape coexistence, which is for example shown by the presence of ground-state shape staggering around the $N = 104$ neutron mid-shell. This staggering was observed by measuring the isotope shift along the chain of isotopes which extended towards the lighter isotopes until mass number $A = 181$. One of the key questions that remained unanswered was where this staggering phenomenon ends.

Several isotopic chains in the lead region that cross the $N = 126$, show a strong deviation from the trend of the lighter isotopes at the neutron shell closure. No such measurements of this ‘kink’ were performed so far in the lead region below $Z = 82$.

The first part of this thesis reports on an in-source laser-spectroscopy experiment performed at the ISOLDE radioactive ion-beam facility in CERN. These were the first measurements of the hyperfine structure and isotopes shifts in the mercury isotopes below $A = 181$ and also the first to go across $N = 126$ on the neutron-deficient and neutron-rich sides of the chain respectively.

The mercury radioactive ion beams were produced by impinging a 1.4 GeV proton beam on a molten-lead target. The reaction products were extracted, resonantly ionized using a three-step laser-ionization scheme, mass separated and sent to one of three detection setups. Strongly-produced ions were measured on a Faraday Cup. The α and γ decay of short-lived and weakly-produced isotopes ($^{177-182}\text{Hg}$) was measured at the Windmill setup with silicon and germanium detectors. These decay modes were used to identify the isotopes of interest from the contaminated ion beam. Isotopes undergoing mainly β decay $^{182-185,202,202,206-208}\text{Hg}$ were identified using their time-of-flight in a Multi-reflection Time-of-Flight mass spectrometer.

From the results, both the end-point of the shape staggering at $A = 180$ and the observation of a charge-radius kink at $N = 126$ are confirmed. The results of the neutron-deficient isotopes were compared to state-of-the-art theoretical calculations using either Density Functional Theory (DFT) or the Monte-Carlo Shell Model (MCSM). The DFT calculations are able to qualitatively reproduce a ground-state staggering by tuning different functional parameters. By calculating the experimentally-observed spin states, the MCSM results reproduce the shape-staggering and electromagnetic moments of the isotopes.

In the second part of this thesis, the design, simulation and commissioning of radio-frequency quadrupole (RFQ) ion guides are discussed. In the commissioning tests of the RFQ ion guides, the transport efficiency, longitudinal emittance, beam energy spread and time profiles were measured. These results were compared to simulations performed with SIMION and IonCool software packages. The ion guides form part of an experimental setup that is dedicated to the development of in-gas(-jet) laser-ionization spectroscopy. This technique makes use of the properties of a supersonic gas jet to produce an ensemble of atoms that is suited for high-resolution laser-spectroscopy studies. Besides this, the technique is also suited for the creation of exotic beams of radioactive ions, when it will be installed at facilities using in-flight production of isotopes.

Beknpte samenvatting

De neutronarme kwikisotopen bevinden zich in het gebied rond de $Z = 82$ proton schilsluiting, die gekend is voor zijn manifestatie van vormcoëxistentie. Vormcoëxistentie is het fenomeen waarin verschillende sterktes of types van kernvorming aanwezig zijn bij lage excitatie-energie. De kwikisotopen zijn zelfs één van de voornaamste voorbeelden van vormcoëxistentie. Dit uit zich bijvoorbeeld in de aanwezigheid van een abrupte vormverandering in de grondtoestand wanneer een neutron uit de atoomkern wordt weggenomen. Meer specifiek gebeurt dit in het gebied rond $N = 104$ in het midden van de neutronschild. De abrupte vormverandering kan worden geobserveerd door het meten van de ladingstraalverschillen langsheen de keten van isotopen. De oneven kernen $^{181,183,185}\text{Hg}$ vertonen een sterk afwijkende grondtoestandsvervorming in vergelijking met hun even burens $^{182,184,186}\text{Hg}$, die slechts licht vervormd zijn in hun grondtoestand. Tot dusver reikten zulke metingen in de neutronarme kwikisotopen tot ^{181}Hg . Eén van de centrale vragen die tot nu toe onbeantwoord bleef, is waar de abrupte vormverandering eindigt. Daarenboven vertonen verschillende isotoopketens in de loodregio een afwijking in hun ladingstraal van de trend die wordt gezet door de lichtere isotopen. De afwijking toont zich als een karakteristieke ‘knik’ in de ladingstraal bij de $N = 126$ schilsluiting. Tot nu toe werden er in de loodregio geen metingen uitgevoerd van deze knik in het gebied onder $Z = 82$.

Het eerste deel van deze thesis beschrijft een in-bron laserspectroscopie-experiment op kwik dat werd uitgevoerd aan de ISOLDE faciliteit in CERN waar radioactieve ionenbundels worden aangemaakt. Het doel van deze campagne was om het gebied van abrupte vormverandering af te lijnen en te bepalen of de ladingstraalknik ook aanwezig is in isotoopketens in de loodregio met $Z < 82$. Door het opmeten van ionizatierezonanties bij het variëren van de laserfotonfrequentie werd de isotoopshift en het hyperfijnspectrum opgemeten. Hieruit werden vervolgens de ladingstraalverschillen en elektromagnetische momenten van de grondtoestanden bepaald. De isotopen werden in de

metingen geïdentificeerd door hun karakteristieke radioactieve α - en γ -verval in de Windmill detectieopstelling of door hun transitietijd in een multireflectie *time-of-flight* spectrometer. Dit experiment was het eerste dat de isotoopshift en hyperfijnspectra in lichte kwikisotopen onder $A = 181$ opmeette en was het eerste dat tot voorbij de $N = 126$ schilsluiting reikte in de neutronrijke kwikisotopen.

Uit deze resultaten werd zowel het eindpunt van de abrupte vormverandering vastgesteld bij ^{180}Hg , als werd voor het eerst een ladingsstraalknik bevestigd bij $N = 126$ in een isotoopketen in de loodregio onder $Z = 82$. De resultaten van de neutronarme kernen werden vergeleken met state-of-the-art theoretische berekeningen van dichtheidsfunctionaaltheorie (DFT) en van het Monte-Carlo schillenmodel (MCSM).

In het tweede deel van deze thesis, worden het ontwerp, de simulaties en de eerste tests besproken van de radiofrequentie quadrupool ionengeleiders. De ionengeleiders maken deel uit van een experimentele opstelling die is toegewijd aan de ontwikkeling van de in-gas-(jet) laserionizatiespectroscopie (IGLIS). Deze techniek maakt gebruik van de eigenschappen van een supersonische gasjet om een ensemble van atomen te creëren dat geschikt is voor hoge resolutie laserspectroscopiestudies. Hiernaast is de techniek ook geschikt voor de aanmaak van radioactieve ionenbundels van exotische isotopen. In de operationele tests werden de transportefficiëntie, longitudinale emittantie, energiespreiding en tijdsprofielen van de ionenbundel opgemeten. De resultaten werden vergeleken met simulaties die werden uitgevoerd met de softwarepakketten SIMION en IonCool dewelke verschillen in het berekenen van de atoom-ion botsingsinteractie. Uit de tests werd een operationeel werkingsdiagramma opgesteld in dewelke de ionengeleiders optimaal werken. In de toekomst zullen vergelijkbare ionengeleiders geïmplementeerd worden in een opstelling aan de nieuwe S^3 faciliteit in GANIL (Caen, Frankrijk), waar de IGLIS techniek zal worden gebruikt voor zowel de studie als de productie van exotische radioactieve ionenbundels. Dit zal het mogelijk maken bepaalde regio's van de kernkaart te bestuderen, zoals de regio's van de (super)zware of de refractaire elementen, die momenteel buiten het bereik liggen van conventionele laserspectroscopiestudies.

Contents

Abstract	v
Contents	ix
List of Figures	xiii
List of Tables	xxi
0 Optical spectroscopy across the nuclear landscape	1
I In-source laser ionization spectroscopy of mercury	7
1 Introduction	8
1.1 Motivation for studying mercury isotopes	8
1.1.1 Shape coexistence in the lead region and shape staggering in neutron-deficient mercury isotopes	9
1.1.2 Charge-radii kink at N=126	11
2 Nuclear Models	13
2.1 Liquid drop and droplet model	14
2.2 The nuclear shell model	15

2.2.1	Spherical shell model	15
2.2.2	Shell model calculations	20
2.2.3	Deformed shell (Nilsson) model	23
2.3	(Beyond-)Mean Field models	26
3	Theoretical basics for laser spectroscopy	29
3.1	Hyperfine structure and nuclear moments	29
3.1.1	Magnetic dipole moment	31
3.1.2	Electric quadrupole moment	31
3.1.3	Hyperfine anomaly	32
3.2	Isotope shift and change in mean-square charge radius	33
3.2.1	Mass Shift	33
3.2.2	Field Shift	34
3.2.3	King plot	34
3.2.4	Mean-square charge radius	35
3.3	Broadening mechanisms of spectral lines	36
3.3.1	Natural line width	36
3.3.2	Doppler broadening and shift	36
3.3.3	Pressure broadening and shift	37
3.3.4	Power broadening	38
4	Laser spectroscopy of mercury isotopes	39
4.1	Experimental setup at ISOLDE, CERN	39
4.1.1	Resonant laser ionization	43
4.1.2	Windmill detection setup	44
4.1.3	MR-TOF-MS	49
4.2	Analysis	51
4.2.1	WM data	51

4.2.2	Reference measurements of ^{198}Hg	57
4.2.3	Fitting	60
4.3	Summary of results and comparison to literature	63
4.3.1	Determination of charge radius difference and deformation	65
4.3.2	Determination of nuclear moments	68
4.4	Publications (Articles I and II)	73
II	An ion-beam cooler and guide for IGLIS	103
5	In-gas-jet laser ionization spectroscopy	104
5.1	In-gas-jet laser ionization and spectroscopy	105
5.1.1	Comparison to other laser spectroscopy techniques . . .	107
5.2	Experimental setup at KU Leuven	108
5.2.1	Jet laboratory	109
5.2.2	Laser laboratory	110
6	Ion guide and beam cooler principles	113
6.1	Ion trapping with RF potentials	113
6.2	Buffer gas cooling	117
7	Simulations and design	119
7.1	Design	120
7.1.1	SRFQ	121
7.1.2	DPRFQ	123
7.1.3	IGRFQ	125
7.1.4	Complete Setup	127
7.2	SIMION and IonCool simulations	127
7.3	Electronics	129

8	Commissioning tests	133
8.1	Discharge tests	133
8.2	Transport tests	136
8.3	Longitudinal E-spread	141
8.4	Time profile measurements	142
8.5	Emittance measurements	147
8.6	Mass scans and contamination	153
9	Conclusions and outlook	159
9.1	Conclusions	159
9.1.1	Laser spectroscopy of mercury	159
9.1.2	RFQ ion guides	161
9.2	Outlook	162
	Bibliography	165

List of Figures

1	Nuclear chart in which isotopes that have been studied using optical spectroscopy are indicated in red. Stable isotopes are shown as black squares. Figure adjusted from [17]	4
1.1	Calculated potential energy surface of ^{186}Pb as a function of nuclear shape parameters β_2 and γ . Different minima are indicated with their corresponding shapes. Figure from [6] . . .	8
1.2	Ground-state mean-square charge-radii differences in the lead region. The atomic number Z is shown for each isotopic chain. Data until 2017 from [28] and [29–33].	10
2.1	Single-particle energies for a simple harmonic oscillator (SHO), an extended harmonic oscillator with an ℓ^2 -term and a realistic shell-model potential with ℓ^2 and spin-orbit ($\ell.s$) terms. Figure adjusted from [59].	17
2.2	Example of the shell-model core and valence space for protons and neutrons used in MCSM calculation of ^{180}Hg described in this thesis. The two-body nucleon-nucleon interactions are indicated by the line and arrows noted $p-p$, $n-n$ and $p-n$ for proton-proton, neutron-neutron and proton-neutron interactions respectively.	18
2.3	Representation of the effect of the tensor force on the relative spacing of different orbitals. Figure from [62].	19
2.4	Experimental excitation energies up to ≈ 3.0 MeV (a) and the theoretical results from IBM-CM calculations (b). Figure from [63]	21

2.5	Dimension of the shell-model calculations with the Lanczos method (filled circles) and the MCSM (blue and red squares) plotted against the year of publication or presentation. The N_{sh} denotes the number of major harmonic-oscillator shells for MCSM calculations without a fixed core as basis. The dashed line is drawn to guide the eye. Figure from [66]	22
2.6	Nuclear shapes corresponding to a spherical, oblate and prolate shape with $\beta_2 = 0$, $\beta_2 < 0$ and $\beta_2 > 0$	23
2.7	Nilsson diagram for neutrons in the range of $82 < N < 126$. Figure from [69]	25
2.8	(top) schematic representation of a $\beta - \gamma$ plane. (bottom) Energy surface for IBM-CM calculations for $^{182-186}\text{Hg}$ (see section 2.2.2). The energy spacing between adjacent contour lines equals 100 keV and the deepest energy minimum is set to zero, corresponding to the red color. Figure from [63].	26
3.1	Hyperfine level structure for ^{179}Hg of the $6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1$ transition. The right-hand side shows the splitting of the 3P_1 level into different F-states for an $I = 7/2^-$ nuclear spin. An exemplary spectrum is given on top where the resonance peaks correspond to the transitions shown by the red arrows below.	30
4.1	ISOLDE layout. Picture adjusted from [89]	40
4.2	Schematic overview of the experimental setup.	41
4.3	Ionization scheme of mercury that was used during the experiment and developed in [90].	43
4.4	Model of the Windmill detection setup. Left: The silicon detector pairs (brown), carbon foils (light grey), rotatable wheel (black) and the Low-Energy germanium detector (LEGe, blue) are shown. The germanium detector positioned at a 90° angle with the beam path is not visible in this sketch. Right: zoomed in view of the implantation site with α particles (orange), β /conversion electrons (blue), γ rays (green) and fission fragments (FF, black) originating from the foil position. Figure from [93]	45
4.5	Calibration curves for germanium (top) and silicon (bottom) detectors.	46

4.6	Top: Schematic data acquisition and signal processing layout. Bottom: Timeline of acquisition with several timing and veto signals superposed on the supercycle structure.	47
4.7	Layout of computers and information flow of the experiment . .	48
4.8	ISOLTRAP's Multi-reflection Time-of-flight (MR-TOF) Mass spectrometer/separators. Picture taken from [94].	49
4.9	Top: Comparison of TOF versus intensity for all mercury isotopes measured with the MR-TOF setup. Bottom: Zoom in of the TOF spectrum of ^{208}Hg . Figures provided by F. Wienholtz. . .	50
4.10	Summary of the mercury isotopes measured in the experiment, indicated by bold frames. Colored squares indicate the detection setup used for scanning. The Windmill, MR-TOF and Faraday Cup setups are indicated by red, green and pink squares respectively. Top: Overview of the total region-of-interest. Bottom: Zoom-in on neutron-deficient (left) and neutron-rich (right) parts of the isotopic chain.	51
4.11	Overview of the alpha spectra obtained with Si detectors at the implantation site, summed for all HFS scans.	52
4.12	LEGe spectrum on/off resonance for a HFS scan of ^{181}Hg . The origin of gamma's with a large on/off resonance count difference are indicated.	53
4.13	HFS scan of ^{181}Hg gating on alpha (black) and 148 keV gamma (red) energies.	54
4.14	Deadtime correction factor for the example of ^{181}Hg , where the deadtime correction was the largest of all scans. Top: The original detected α counts as a function time during the laser-frequency step (y-axis) and in function of start-time of each laser step (x-axis). Middle: The fraction of recorded pulses of a 100 Hz pulser in function of time. Bottom: The same spectrum as shown on top, but now corrected for using the pulser-correction factor.	56
4.15	Comparison of the uncorrected (red) hyperfine spectrum of ^{181}Hg with the deadtime-corrected spectra obtained with methods 1 (black) and 2 (blue).	57
4.16	Summary of the timing of all scans performed for different mercury isotopes relative to the start of the experiment. The symbols indicate the method used for obtaining the scan. . . .	58

4.17	Example spectrum of ^{198}Hg Faraday Cup reference scan. . . .	59
4.18	Tripled centroid frequencies of ^{198}Hg reference measurements with respect to the weighted average of the FC measurements for all FC and MR-TOF scans. MR-TOF scan number 2 is omitted due to being incomplete.	59
4.19	Top: Example fit for a HFS scan of ^{179}Hg . Bottom: The error analysis performed with SATLAS using the EMCEE Markov-chain Monte-Carlo method. The probability distributions for the (shifted) centroid, hyperfine a and b parameters are presented on the diagonal histograms. The 2-dimensional off-diagonal plots show the correlation between different parameters and the 1σ , 2σ and 3σ contours. Values are quoted in fundamental (non-tripled) frequencies.	61
4.20	Summary of fits for all isotopes measured in this experiment. The red lines show the fit results of the data indicated by the black markers. In case of ^{185}Hg , the additional green (blue) line shows the partial fit result of the ground (isomeric) state. . . .	62
4.21	Overview of mercury charge radii differences obtained from this work and from [5]	66
4.22	Overview of mercury charge radii differences around the $N = 126$ shell closure obtained from this work and from [5]. The blue line was added to guide the readers eye.	67
4.23	Overview of mercury magnetic moments and g -factors from this work and from [5]. The theoretical g -factor Schmidt values ($g_{\text{eff}} = 0.6g$) for odd-neutrons are shown by dashed lines.	69
5.1	Cut view of the Leuven gas cell with the main components indicated. Figure adjusted from [120]	105
5.2	Comparison of in-gas-jet and in-gas-cell laser ionization spectroscopy of actinium isotopes studied at LISOL. Figure from [13]	107
5.3	Layout of the IGLIS laboratories at KU Leuven. Figure from [120]	108
5.4	Top: Inside layout of the jet lab frontend at KU Leuven. Bottom: Picture of the actual setup.	109
5.5	Picture of the HV section of the jet laboratory with the main components indicated.	110

5.6	Picture of the laser laboratory clean room (ISO8) with most important components indicated.	111
6.1	Left top: Rod configuration for producing a radially confining electric field using RF potentials. Left bottom: Time dependence and relative phase of the applied potentials. Right: Representation of the saddle potential created by applying potentials 180° out of phase to adjacent rods at time t	114
6.2	Simulated motion of Cu^+ ions in a linear RFQ showing the large-amplitude macromotion and small-amplitude micromotion superimposed. The effect of the axial DC dragging field is seen as the increase in axial movement per cycle (indicated by the black arrows).	115
6.3	Mathieu stability diagram for ion motion as a function of the stability parameters a, q given in equations 6.6 and 6.7. Figure from [123].	116
7.1	Frontend design with different pressure regimes.	119
7.2	Vacuum calculations diagram.	120
7.3	Design of the SRFQ.	121
7.4	Relationship between jet diameter and Doppler width for supersonic jets of different Mach numbers. Figure taken from [120]	122
7.5	Photograph of the SRFQ ion guide.	123
7.6	Transmission efficiency of the DPRFQ versus inner radius r_0	124
7.7	Picture of the DPRFQ ion guide.	125
7.8	Simulated effect of an extraction plate on the penetration of the electric field inside the final IGRFQ section. Contour lines differ 1, 10 and 50 V for consecutive red, blue and green lines respectively. The simulations shown here were performed with SIMION (Sec. 7.2).	126
7.9	Photograph of the IGRFQ ion guide.	126
7.10	Photograph of the complete setup of ion guides installed in the IGLIS frontend vacuum chamber.	127

7.11	Longitudinal velocity profile of Mach 8 jet as simulated by COMSOL. COMSOL calculations provided by E. Mogylevsky.	128
7.12	Interaction potentials for ion-buffer gas collisions.	129
7.13	Layout of RFQ electronics.	130
7.14	Resonance circuit for generation of the RF voltages.	131
8.1	Pashen curves for several common gas types. Parameters used for calculation from [138].	134
8.2	Example of an electrical discharge induced in the SRFQ.	135
8.3	Breakdown voltages as a function of buffer-gas pressure in the SRFQ.	135
8.4	Transport measurement method.	136
8.5	Ionization scheme of copper used for resonant ionization.	137
8.6	Example of a transport measurement performed with laser-ionized copper isotopes.	138
8.7	Total transmission efficiencies of $^{63,65}\text{Cu}$ ions measured at different pressures in the jet chamber compared to simulations performed with SIMION and IonCools.	139
8.8	Operational diagram of the setup. The RFQ-transport limitations (horizontal solid black lines) are combined with the optimal jet-formation pressures in the jet chamber for different Mach numbers and stagnation pressures P_0 . The optimal operational regime is shown in light green. Downwards-pointing arrows delineate the Mach-number range that can be probed for $P_0 = 100, 250$ and 500 mbar shown in solid blue, dashed black and solid red lines respectively.	140
8.9	Bottom: measurement of the energy spread by varying the transmission as a function of blocking voltage applied at the second pumping barrier. The blocking voltage is measured with respect to voltage applied on the first IG electrode. Top: simulation of the experiment using IonCool.	141
8.10	Measurement of the energy spread for different pressures.	142
8.11	Time profile measurement method.	143

8.12	Normalized (peak value = 1) time profiles of ions created using in-gas-cell ionization (setup explained in text) and in-gas-jet ionization.	144
8.13	Time profiles obtained when changing the SRFQ voltage gradient.	144
8.14	Transport times of ^{63}Cu at different pressures and voltage gradients across the SRFQ. Voltage gradients are indicated in the top left corner of each panel. In order to provide a fair comparison of the time profiles, since the filament current and laser power varied for the different tests, the maximum of their value of their time profile was all normalized to 1.	145
8.15	Comparison of experimental and calculated transport times using IonCool, with either the hard sphere (HS) or realistic potential (RP) models.	146
8.16	Emittance measurement setup.	148
8.17	Top: Example of a beam spot recorded at the MCP positioned before the dipole magnet at an einzel lens voltage of 27400 V at a jet-chamber gauge pressure of 4×10^{-2} mbar and 32 V SRFQ gradient. Bottom: Beam spot size as a function of einzel lens voltage with fit results shown in red.	149
8.18	IonCool simulated emittance of ^{63}Cu at the MCP position, for the same pressure as in Figure 8.17, with a SRFQ gradient of 32 V.	150
8.19	Left: Example of a beam spot suffering from strong aberrations measured at the MCP detector for extraction electrode and einzel lens voltages of 400 and 26000 V respectively. Right: Effect of steering causing the beam to only partially hit the MCP detector at higher einzel lens voltages.	151
8.20	Recorded beam sizes for different pressures inside the SRFQ. . .	151
8.21	Simulated emittances for ^{63}Cu at the MCP position for different pressures with a 22 V SRFQ gradient. Simulations were performed with the realistic potential model in IonCool.	152
8.22	Left: Mass scans obtained with both in-gas-cell laser ionization before and after conditioning of the gas cell in linear (1a) and log scale (1b). Right: Mass scans obtained with in-gas-cell (2a) and in-gas-jet (2b) laser ionization both taken before conditioning of the cell shown in linear (2a) and log scale (2b). Labels indicate the most likely origin of the copper adducts.	154

- 8.23 Top: Time profiles for different ^{63}Cu adducts produced via in-gas-cell ionization measured after mass separation. Bottom: Zoomed-in version of the same data as presented in the top plot. 157

List of Tables

4.1	Setup, measurement time and corresponding maximum event rate for the isotopes of interest on resonance during HFS scans of laser-spectroscopy experiment IS598. For the windmill measurements, the obtained α rate is given for both silicon detectors at the implantation site. For MR-ToF measurements, the production rate is given in counts/shot.	42
4.2	Summary of α -tagging contributions to the HFS measured with the WM setup and their corresponding half-lives, alpha-particle energies and intensities. Data from [95].	55
4.3	Reference scans used in this study for the IS determination. . .	63
4.4	Comparison between IS results obtained in this analysis and that of A. Barzakh.	64
4.5	Comparison between HFS a -factor results obtained in this analysis and that of A. Barzakh.	64
4.6	Comparison between HFS b -factor results obtained in this Analysis and that of A. Barzakh.	65
4.7	$\langle\beta^2\rangle^{1/2}$ -deformation parameters extracted from the charge-radii as given further in Table 4.10, normalized to ^{198}Hg with deformation parameter $\langle\beta^2\rangle^{1/2} = 0.1102(7)$	68
4.8	BW and BR contributions to the hyperfine anomaly	69
4.9	Summary of isotope shifts and hyperfine parameters for $6s6p^3P_1$ level in mercury and their comparison to literature.	71
4.10	Summary of resulting mean-square charge differences ($\delta\langle r^2\rangle^{A-198}$) and nuclear moments (μ, Q) and their comparison to literature.	72

- 8.1 Summary of Cu-adducts formed with impurities present in the gas and their dissociation energy. Data from [144] and [136] . 155

Part 0

Optical spectroscopy across the nuclear landscape

-the link between parts I and II-

The drive towards higher sensitivity and spectral resolution

In this chapter, the link between parts I and II of this thesis is detailed. While the first part of this work deals with the experimental study of mercury using laser spectroscopy at the ISOLDE facility in CERN, the second part discusses the design and commissioning of an ion-beam cooler and guide as part of a dedicated setup at KU Leuven for developing the in-gas(-jet) laser ionization spectroscopy (IGLIS) technique.

The nuclear landscape is filled with exciting and fascinating nuclear-structure phenomena. Examples of these phenomena include the presence of different exotic nuclear shapes such as e.g. pear-shaped [3] or halo nuclei [4] as well as rapidly changing nuclear deformation in shape-staggering [1, 5] or shape coexistence [6] and the disappearance of classical and rise of new magic numbers [7]. These phenomena arise from the subtle interplay of single-particle and collective degrees of freedom between nucleons comprising the nucleus [8]. While the underlying forces that govern the interaction between quarks are well described in the Standard Model by Quantum Electro Dynamics (QED) and Quantum Chromodynamics (QCD), a full description of the nucleon interaction and nuclear-structure phenomena starting from such basic principles is lacking. Lattice QCD calculation attempts to provide such a description are promising but unfortunately so far limited to only the lightest of elements with $A < 5$ [9]. Theories such as Chiral Effective Field Theory (χ EFT) are being developed that are able to provide nuclear interaction potentials using baryon and meson degrees of freedom without loss of consistency with chiral symmetries of QCD [10]. Ab initio theories are now reaching mid-mass nuclear systems even for open-shell nuclei [11]. For heavier systems, such models are currently unavailable

and one has to rely on theories making use for instance of phenomenological interactions fitted to different observables across the nuclear chart. However, theories for these heavier systems are continuously refined, allowing for larger and more accurate calculations. Recent advances include for instance the development of more refined interaction potentials fitted for a wide range of nuclear observables such as e.g. UNEDF1 [12]. Another breakthrough in theoretical calculations for heavier systems is the development of the Monte-Carlo shell model (MCSM), where many nucleons interacting in large model spaces can be calculated by making use of Monte-Carlo techniques to avoid a currently impossible diagonalization of large Hamiltonians.

In order to test the validity and accuracy of such new advances in nuclear theory, there is a need for experimental benchmarks. Such benchmarks come from nuclear observables usually measured to high accuracy. Nuclear-structure observables such as excitation energy spectra, electromagnetic transition strengths, nuclear masses, charge radii, spins and electromagnetic moments are part of a broad scope of observables available for comparing theoretical calculations with experimental results. Benchmarking nuclear models for exotic nuclei with extreme neutron-to-proton ratios, different from stable isotopes, is desired as certain features of the nuclear force are amplified and more prominent than next to stability. This results in a continuous demand to push the limits of accuracy and sensitivity of current experiments trying to measure these observables in order to not only test the reach of current and new nuclear theories but even answer fundamental questions such as "Where does the nuclear chart end?". In laser spectroscopy experiments for example, in-source laser spectroscopy is extremely sensitive, being able to measure isotopes with a production rate of <0.1 atoms/second, as shown in this thesis, but lacks the spectral resolution provided by collinear laser spectroscopy (CLS) to measure optical transitions down to their natural linewidth and vice versa. Therefore, several different approaches are currently being pursued to combine high sensitivity with high spectral resolution [13–16]. Such a technique would allow for a precise measurement of the hyperfine spectrum and isotopes shifts of the most exotic or heaviest nuclear isotopes and as such provide an excellent tool for validating new theoretical advances through the measurement of charge radii differences, spins and electromagnetic moments.

In Figure 1, the nuclear chart is shown on which isotopes that have been studied using optical spectroscopy are indicated by red squares. Stable isotopes are indicated in black, and green squares denote isotopes that were already studied but not yet published at this thesis publication date. Large regions of this map are present for which experimental data is scarce or non-existent. A few examples include regions of refractory elements, for which extraction from

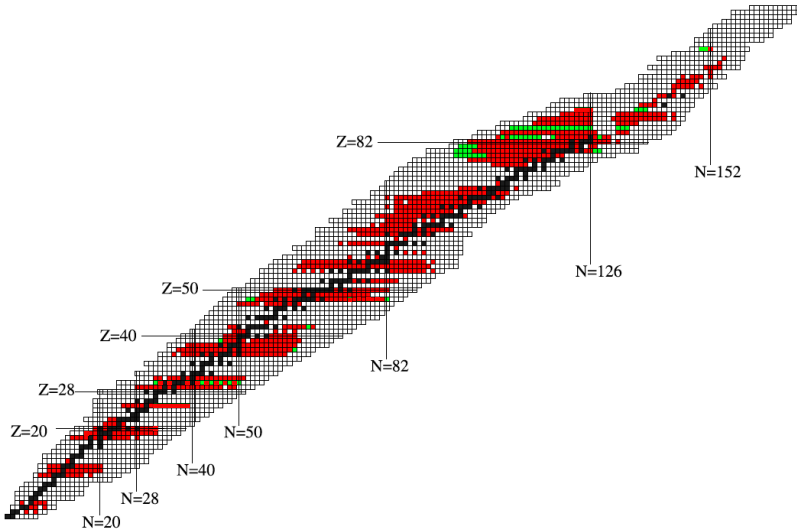


Figure 1: Nuclear chart in which isotopes that have been studied using optical spectroscopy are indicated in red. Stable isotopes are shown as black squares. Figure adjusted from [17]

thick ISOL-type targets is difficult [18] or the heavy element region, for which production rates are extremely low due to the very small reaction cross sections ($\leq \text{nbarn}$) [19, 20] and which cannot be produced by high-energy proton-induced reactions. Most of these measured isotopic chains do not extend all the way to the proton or neutron drip lines. Limitations here are posed on the one hand by the very short lifetimes associated with nuclei far from stability and on the other hand by the low production rates in comparison to neighbouring masses and higher production of isobaric contaminants in the ion beam.

This work is largely split in two parts. On the one hand, it deals with laser spectroscopy of mercury (Hg) isotopes studied at ISOLDE, CERN described in Part I. Here, the sensitive technique of in-source laser spectroscopy is combined with alpha-decay spectroscopy and Time-of-flight mass separation providing nearly background free hyperfine spectra. This allowed to extend previous Hg laser spectroscopy measurements closer to the proton dripline and further across the $N = 104$ midshell region. On the other hand, this work concentrates on the design, simulation and commissioning of an ion-beam cooler as part of a new laboratory dedicated to in-gas(-jet) laser ionization spectroscopy (IGLIS) developments, described in Part II. The construction and commissioning of part of the IGLIS setup at KU Leuven contributes to reaching the long-term goal of

performing laser spectroscopy in the refractory and heavy element regions of the nuclear chart. Although both parts of this thesis might appear unrelated at first, their connection lies in the drive to push laser spectroscopy to the limits of what is currently possible in terms of sensitivity and spectral resolution.

Part I

In-source laser ionization spectroscopy of mercury

Chapter 1

Introduction

1.1 Motivation for studying mercury isotopes

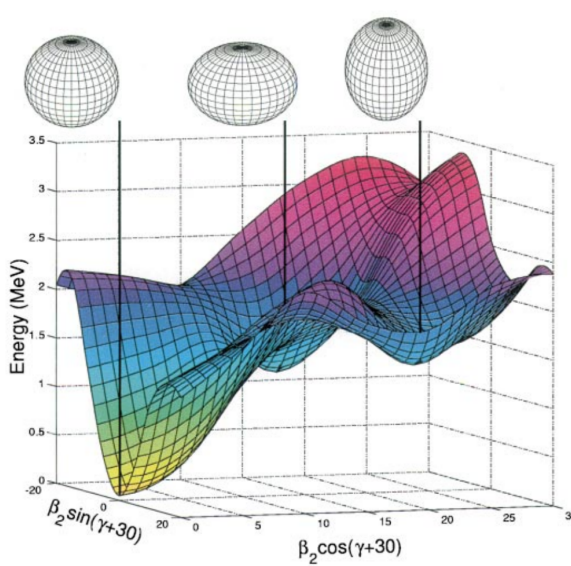


Figure 1.1: Calculated potential energy surface of ^{186}Pb as a function of nuclear shape parameters β_2 and γ . Different minima are indicated with their corresponding shapes. Figure from [6]

Shape coexistence refers to the presence of one or more low-lying excited nuclear states with different shapes compared to the nuclear ground state. A famous example is the case of ^{186}Pb shown in Fig. 1.1, where three different 0^+ -states, all within 1 MeV, are assumed to exhibit spherical, oblate and prolate shapes [6].

The origin of shape coexistence lies in the competition between two opposing tendencies being on the one hand a stabilizing effect of closed (sub)shells favouring spherical shapes and on the other hand residual nucleon-nucleon interactions driving deformation [8].

To this day, mercury remains one of the prime examples of shape coexistence and one of the most-studied isotopic chains in the lead region [21]. However, several long-standing open questions remain unanswered. The first question relates to the so-called shape staggering of the ground-state charge radius in neutron-deficient mercury isotopes, first observed through optical spectroscopy measurements at ISOLDE [22]. Secondly, beyond $N = 126$, a kink in the charge-radius trend is observed for several isotopic chains in the lead region above $Z = 82$ such as radium ($Z = 88$), francium ($Z = 87$), radon ($Z = 86$), astatine ($Z = 85$), polonium ($Z = 84$), bismuth ($Z = 83$) and lead ($Z = 82$) [21, 23–27]. Measurements of the charge radius of mercury have so far only been done for isotopes with $181 \leq A \leq 206$ [5]. Extending these measurement would answer the questions such as "Where does the shape staggering end?" and "Does the kink in $\delta\langle r^2 \rangle$ at $N = 126$ also extend south of the $Z = 82$ closed shell?". The following sections 1.1.1 and 1.1.2 give a more detailed explanation of these questions which will be addressed in this thesis.

1.1.1 Shape coexistence in the lead region and shape staggering in neutron-deficient mercury isotopes

Review papers refer to the neutron-deficient lead region as the region of the most extensive manifestation of shape coexistence on the nuclear chart [8, 17, 34]. Within microscopic shell models detailed in section 2.2, shape coexistence arises from particle-hole excitations across a shell gap when the different nucleon-nucleon interactions render this energetically favourable. Mean-field descriptions (Sec. 2.3) explain shape coexistence as the emergence of several local minima with different deformation parameters in the global potential energy surfaces of nuclei.

Shape staggering in the lead region was first discovered in neutron-deficient mercury isotopes [22] by measuring isotope shifts using a technique named β -radiation detection of optical pumping (β -RADOP) and has been called "one of the most remarkable discoveries in nuclear structure physics in the last 50

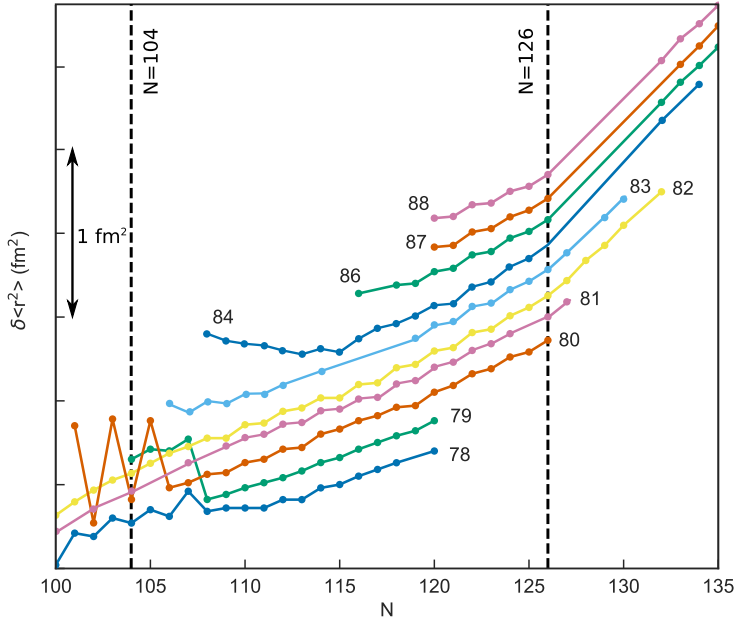


Figure 1.2: Ground-state mean-square charge-radii differences in the lead region. The atomic number Z is shown for each isotopic chain. Data until 2017 from [28] and [29–33].

years" [34]. Using β -RADOP, a technique only suitable for measuring nuclei with non-zero nuclear spin and thus only odd-mass isotopes of Hg, Bonn et al. found a large isotope shift between $^{183,185}\text{Hg}$ and ^{187}Hg indicating a transition from an assumed small oblate to large prolate nuclear deformation. Not long after these first measurements, in-beam γ -ray spectroscopy could not confirm similar deformation for their even-mass neighbors $^{184,186}\text{Hg}$ that appeared to be weakly oblate deformed [35, 36]. It thus became clear that the shapes of neutron-deficient mercury isotopes staggered between small and large nuclear deformations from one isotope to the other. Soon thereafter, the group of Bonn also found that ^{181}Hg exhibits the same behaviour as $^{183,185}\text{Hg}$ [37]. As optical spectroscopy developed and lasers started playing an important role in the field, even-mass mercury isotopes could also be measured with this technique. A series of laser spectroscopy measurements followed from 1977 through 1986 [5, 38, 39] and brought the knowledge of isotope shifts, hyperfine parameters and from that, charge radii and electromagnetic moments. This is how the situation remained for more than 30 years. A plot of the ground-state charge radii in

the lead region, also showing mercury ($Z = 80$) measured until then, is shown in Fig 1.2. The lightest mercury isotope measured with optical spectroscopy was still ^{181}Hg and a direct measurement of the endpoint of shape staggering remained absent. This work builds upon this first finding and all subsequent optical spectroscopy measurements with the goal of finding the endpoint of the shape staggering phenomenon in mercury by measuring more neutron-deficient mercury isotopes than has been done so far.

1.1.2 Charge-radii kink at $N=126$

Besides the clear shape staggering in mercury around the $N = 104$ mid-shell shown in Figure 1.2, a characteristic change in the slope is observed in the trend of the charge radii differences, $\delta\langle r^2 \rangle$, at the neutron shell closure $N = 126$. This kink is also present at other neutron shell closures e.g. $N = 50, 82$ [28] and the magnitude of the gradient change seems to decrease with proton number Z [17]. The droplet model, an often used model to describe charge radii differences in the lead region, qualitatively describes a smooth $\delta\langle r^2 \rangle$ -behaviour as a function of N and cannot account for the observed shell effects. Even for more advanced descriptions, such as (relativistic) mean-field models, predicting the presence and magnitude of the kink remains a challenge [17, 40–42] and no general consensus exists on how to reproduce the correct magnitudes [26]. The isotopes with $N = 127$ and 128 close to $Z = 82$ are particularly difficult to access and data are scarce [43]. Below the $Z = 82$ shell closure, there is a complete lack of data, as can be seen from Figure 1.2. Therefore, it is not known if the kink will be present below the $Z = 82$ shell closure. Measuring mercury isotopes with $A \geq 206$, and hereby proving the existence and quantifying the strength of the charge-radius kink, would provide data to benchmark current theoretical models and give deeper insight in the underlying mechanism.

Chapter 2

Nuclear Models

The feed-back loop connecting theory and experiment in which theory predicts experimental outcomes and experimental results are used to benchmark and refine theory is a constant drive in the advance of science in general. The ultimate goal of nuclear theory to describe all nuclei and nuclear phenomena across the nuclear chart from their most basic constituents is so far still unreachable. Currently, there is not a single nuclear theory that is capable of describing nuclear behaviour for all regions of the nuclear chart, and several different models are being utilized. These models try to explain and predict observables that can be tested by experiments.

Ab-initio theories, such as e.g. chiral effective field theories, started with perturbative calculations consistent with quantum chromodynamics (QCD) for the lightest nuclei, and are slowly but surely working their way up towards medium-mass regions of the nuclear chart. However, several limitations hinder the path towards the heavy nuclei [44]. Opposed to this ultimate microscopic view, building the nuclear chart bottom up, are macroscopic models such as e.g. the (shell-corrected) liquid drop model, that describe very well average global trends across the nuclear chart [45].

In this chapter, the theoretical models most relevant for this work are discussed. While the liquid drop and droplet model are macroscopic models, both shell-models and mean-field models described in this chapter lie in between these two microscopic and macroscopic extremes.

2.1 Liquid drop and droplet model

The liquid drop model (LDM) is a macroscopic model that describes several nuclear properties. In analogy to an incompressible liquid drop, it models nuclei assuming uniform nucleon densities and a sharp nuclear surface of radius $r = r_0 A^{1/3}$ (where $r_0 = 1.2$ fm). A big achievement of the LDM was that it could reproduce the experimental trend observed in nuclear binding energies $B(Z, N)$ according to the semi-empirical von Weizsäcker formula as followed.

$$B(Z, N) = a_v A - a_s A^{2/3} - a_c Z(Z-1)A^{-1/3} - a_{\text{sym}} \frac{(A-2Z)^2}{A} + \delta \quad (2.1)$$

Five different energy terms related to nuclear volume (a_v), surface (a_s), Coulomb (a_c), symmetry (a_{sym}) and pairing effects (δ) are used to describe the binding energy of a nucleus [46]. The volume term takes into account the short range attractive nature of the strong nuclear force. Since nucleons at the surface have less nearest neighbours than those at the center of the nucleus, they should therefore feel less of the strong binding effect. The so-called surface term takes this into account by reducing the binding energy accordingly. The repulsive Coulomb effect of the charged protons reduces the binding energy as well. The fourth term referred to as the symmetry term favours nuclei with similar proton and neutron numbers in contrast to nuclei with a large proton-neutron number asymmetry. The final term takes pairing effects into account and only adds to the binding energy for even-even nuclei. For odd-A nuclei, the pairing term is zero, and it is negative in odd-odd cases.

The LDM was later refined, at first by relaxing the uniform nucleon density approximation and adding surface diffuseness [47], and was a few years hereafter expanded to include arbitrary nuclear shapes [48]. The refined version was named the Droplet Model (DM).

Currently, the DM is often used to compare charge-radii measurements with predicted trends for different degrees of deformation according to equation 3.20 in section 3.2.4 of the next chapter [17, 23, 29, 49, 50]. Two parametrizations for calculating charge radii in the DM model are given by Myers and Schmidt [51], and by Berdichevsky and Tondeur [52]. The latter increases the slope of the charge-radii prediction by 10-15 % in the lead region [53]. Within the present work, the most-used parametrization of Berdichevsky and Tondeur will be applied in the calculation of charge radii. Further improvements of the model into the so-called Finite-Range Droplet Model (FRDM) leave the predictions for isotope shifts and charge radii almost unchanged with respect to the droplet model [54].

2.2 The nuclear shell model

Experimental evidence for increased stability at certain proton and neutron numbers resulted in the idea that nucleons are arranged in a shell structure similar to that of atomic shells [55]. Around nucleon numbers 2, 8, 20, 28, 50, 82, 126..., data from nucleon-separation energies, excitation energies of first excited 2^+ levels and nuclear masses for example show clear evidence of this enhanced stability [56–58]. In 1948, Goeppert-Mayer and Jensen were the first to describe this shell structure within the so-called nuclear shell model. Now, 70 years later, more advanced versions of this model are still being used to explain and predict nuclear observables.

2.2.1 Spherical shell model

Describing a nucleus, starting from an A -nucleon problem with a two-body potential, the general Hamiltonian is written as

$$H = \sum_{i=1}^A T_i + \frac{1}{2} \sum_{i,j=1}^A V_{i,j} \quad (2.2)$$

where T_i is the kinetic energy of nucleon i and $V_{i,j}$ is the two-nucleon interaction between nucleons i and j . Since no exact description of the interaction potential $V_{i,j}$ is known, a mean-field approximation is introduced. In this approximation, the nucleons are described as moving in an average central one-body nucleon potential $U(r_i)$ created by all nucleons. Under this assumption, equation 2.2 can be rearranged as

$$H = \underbrace{\sum_{i=1}^A (T_i + U(r_i))}_{H_0} + \underbrace{\frac{1}{2} \sum_{i,j=1}^A V_{i,j} - \sum_{i=1}^A U(r_i)}_{H_{\text{res}}} \quad (2.3)$$

When tackling this many-body problem, the aim is to determine $U(r_i)$, so that the last term H_{res} , the residual interaction, can be treated as a small perturbation to the Hamiltonian H_0 [57, 59]. The solutions to the Schrödinger equation with H_0 are the nucleon single particle energies (SPE) [60], such as the ones shown in Figure 2.1 where they are calculated for three different nuclear potentials: a simple harmonic oscillator, an extended harmonic oscillator with an added ℓ^2 term and a realistic potential including ℓ^2 and spin-orbit $\ell \cdot s$ terms. The orbitals in Fig. 2.1, characterized by shell numbers N , orbital angular momentum ℓ and spin $j = \ell \pm s$, are grouped in shells and large gaps appear at

specific nucleon numbers, when each orbital is being filled with its maximum of $(2j + 1)$ nucleons. The nucleon numbers at which these large energy gaps appear (2,8,20...) are called ‘magic numbers’ and nuclei with both such proton and neutron numbers are referred to as ‘doubly-magic nuclei’. The representation in Fig. 2.1 shows the necessity of including the ℓ^2 and spin-orbit terms to reproduce the experimental magic numbers.

Nuclear ground states most often refer to configurations where all nucleons fill the lowest orbitals from the bottom up to the Fermi surface. Nuclei can be excited by moving nucleons (or pairs of nucleons) to higher-lying orbitals. The non-filled nucleon positions resulting from this excitation to higher orbitals are called holes, and are treated the same way as normal particles within the shell model. Completely-filled shells are so-called closed shells.

Nuclei with nucleons outside of a closed shell are often described as nuclei consisting of a core (the closed shells) and valence nucleons residing in orbitals above the core. Shell-model calculations of nuclei usually start from such a fixed core and let the valence nucleons interact within a certain valence space, the set of orbitals which are taken into account in that certain calculation. The residual two-body nucleon-nucleon interaction $V_{i,j}$ depends in a complex fashion on the mean field $U(r_i)$ and on the valence space available. It is therefore referred to as an ‘effective’ interaction [61]. This interaction, expressed using two-body matrix elements, may be derived in an effective way from a fully-microscopic approach using realistic forces, but may also be of a more phenomenological origin, fixed from fitting energy eigenvalues to experimental energies [7, 8]. An example of the model space used for Monte-Carlo Shell-Model calculations (MCSM, see sec 2.2.2) of ^{180}Hg described in this thesis is shown in Figure 2.2.

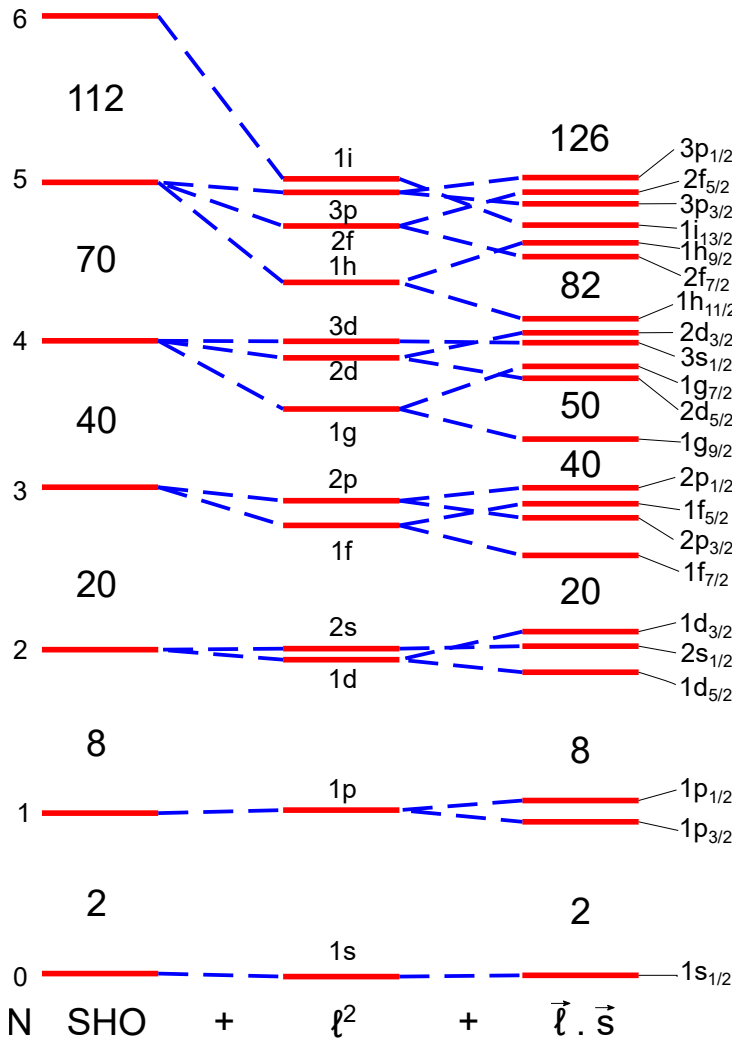


Figure 2.1: Single-particle energies for a simple harmonic oscillator (SHO), an extended harmonic oscillator with an ℓ^2 -term and a realistic shell-model potential with ℓ^2 and spin-orbit ($\ell.s$) terms. Figure adjusted from [59].

A good approximation for the residual interaction in H_{res} of equation 2.3 contains terms involving a pairing interaction and a sum of separable two-body interactions acting between all nucleons, which contains contributions of monopole, quadrupole and higher-order terms. The monopole interaction

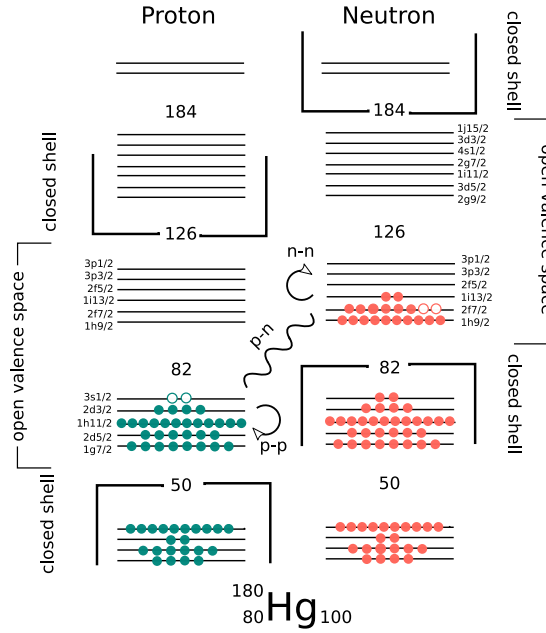


Figure 2.2: Example of the shell-model core and valence space for protons and neutrons used in MCSM calculation of ^{180}Hg described in this thesis. The two-body nucleon-nucleon interactions are indicated by the line and arrows noted $p-p$, $n-n$ and $p-n$ for proton-proton, neutron-neutron and proton-neutron interactions respectively.

has the primary effect of changing SPE's of the nuclear mean field and can therefore even change level-ordering and induce changes in magic numbers. The multipole part of the residual interaction on the other hand, governs a.o. the mixing of shell-model configurations which have the same spin [61]. An effect of such configuration mixing is that nuclei might develop deformation, even within the framework of the spherical shell model, making use of a spherical nuclear potential, as it yields a mechanism that allows for a non-uniform distribution of magnetic m states [59].

The monopole part is described as

$$V_{j,j'}^{\text{mon}} = \frac{\sum_J (2J+1) \langle j, j'; J | V | j, j'; J \rangle}{\sum_J (2J+1)} \quad (2.4)$$

where j and j' are shorthand notation for (n, ℓ, j) of the orbit, $\vec{J} = \vec{j} + \vec{j}'$

and the sum runs over all orbitals in the major shell [60, 62]. The monopole interaction can have the effect of changing the energy of orbitals (e.g. $\pi h_{9/2}$ and $\nu i_{13/2}$ play an important role for the neutron-deficient mercury isotopes), when these orbitals are being filled, thereby effectively decreasing the proton $Z = 82$ shell gap and increasing the probability of exciting protons across this gap. The monopole interaction itself is subdivided in a central, spin-orbit and tensor part [7]. The effect of the central part is increased when the nucleons have the same ℓ values and is in general the strongest term of the monopole interaction. While the spin-orbit part is smaller than the central-part, the difference in effect between spin-orbit partners can become larger even than that of the central part. The tensor part is attractive when the intrinsic spins of the neutron and the proton are anti-parallel ($j_> - j'_<$, for $j_> = \ell + 1/2$ and $j_< = \ell - 1/2$) and is repulsive when they are parallel ($j_> - j'_>$ or $j_< - j'_<$). The effect of the tensor part can be strong enough to have a significant effect on shell evolution [7, 62]. A schematic representation of the effect of the tensor force is given in Figure 2.3.

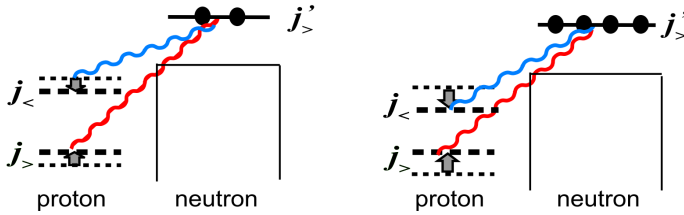


Figure 2.3: Representation of the effect of the tensor force on the relative spacing of different orbitals. Figure from [62].

The pairing term of the residual interaction gives rise to pairing correlations. Its attractive nature is effective for two identical nucleons coupled into pairs with anti-aligned spins adding up to total spin zero ($J = 0^+$) when they have extremely high spacial overlap. It is not limited to diagonal matrix elements acting on pairs within one orbital $\langle j_1^2 0^+ | V_{\text{pair}} | j_1^2 0^+ \rangle$, but allows for scattering of pairs into other orbitals as well $\langle j_1^2 0^+ | V_{\text{pair}} | j_2^2 0^+ \rangle$ [59].

In general, the residual interaction including its pairing, monopole and multipole terms changes the relative energies and/or ordering of the independent particle orbitals of H_0 . Together with this effect, it can account for excitations of nucleon pairs even across shell gaps, creating so-called intruder states. Its induced configuration mixing allows for the development of nuclear deformation.

2.2.2 Shell model calculations

The vast majority of nuclei have nucleon numbers away from only few nucleons above or below closed shells. While shell-model calculations for multi-nucleon configurations are possible, the size of the matrices in which the residual interaction has to be diagonalized, rapidly becomes enormous and intractable [59]. The complexity scales with the amount of valence nucleons that are added to the model space. Truncation of the shell model (space) or different approaches have to be developed to overcome this difficult problem. One has to restrict the model spaces, keeping the essential physics content, if one intends to progress and reach the tin and lead-region nuclei [8].

Interacting boson model

The Interacting Boson Model (IBM) is an attempt to describe heavy nuclei away from closed shells for which the large number of valence nucleons complicate calculations, making use of symmetry arguments. Computational possibilities hamper a full microscopic shell model treatment for such large number of valence particles e.g. around mid-shell, together with valence proton holes and particle-hole excitations across a shell gap [63]. In order to limit the possible nucleon configurations, the IBM model space is truncated by only considering nucleon pairs which are treated as spin $J = 0$ or $J = 2$ bosons. This truncation reduces the computational demand in such a way that calculations for e.g. mercury, platinum, polonium and radon around neutron mid-shell become possible.

For mercury, IBM calculations with the added possibility of configuration mixing (IBM-CM) were recently performed by Garcia-Ramos and Heyde [63] and by Nomura and co-workers [64]. Both use different approaches to the problem. The latter determines the parameters of the IBM Hamiltonian from a mapping of the total-energy surface, derived from self-consistent Hartree-Fock Bogoliubov (HFB) calculations using a Gogny energy functional onto the corresponding IBM mean-field energy (see also sec. 2.3). Garcia-Ramos and Heyde on the other hand fix the Hamiltonian parameters through a least-squares fit to the known excitation energies and $B(E2)$ transition rates of states up to 3 MeV, thereby also allowing for an independent comparison to mean-field calculation results. These calculations seem to describe a number of experimental observables very well such as the excitation spectrum, $B(E2)$ reduced transition probabilities and even-mass charge radii. The excitation spectrum, normalized to ^{198}Hg , is shown in Figure 2.4. In this picture, the excitation energy of a deformed intruder band, built on top of the second 0^+ state, displays a characteristic parabolic

shape with a minimum around $A = 184$. This minimum occurs around neutron number $N = 104$, almost exactly at the middle between two closed neutron shells: $N = 82$ and $N = 126$.

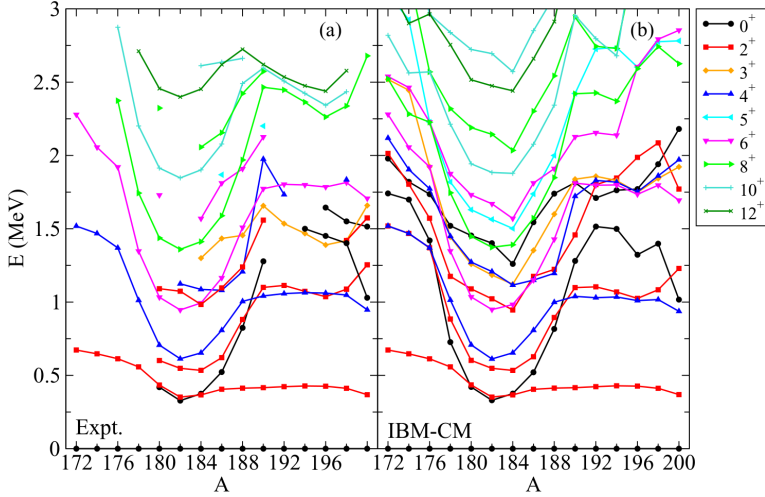


Figure 2.4: Experimental excitation energies up to ≈ 3.0 MeV (a) and the theoretical results from IBM-CM calculations (b). Figure from [63]

Using the parametrization obtained in Ref. [63], a potential energy surface (PES) was extracted which was very similar to the one obtained with the mean-field calculation performed in Ref. [65]. These calculations indicated that, in contrast to experiment, the prolate energy minimum becomes lower than the oblate minimum for $180 \leq A < 186$ (see also Fig. 2.8), being degenerate with the oblate one in $A = 186$ and moving quickly away for $A > 188$. This calculation however, was limited to even-even nuclei and does therefore not provide details of the calculation of the shape staggering in the mid-shell region between $^{181-185}\text{Hg}$.

Monte-Carlo shell model

Conventional shell model calculations are performed by diagonalizing the Hamiltonian matrix of which the dimensions depend on valence orbits and valence nucleons [66]. As stated earlier, the dimensionality of this calculation runs out of hand very quickly. The approach of the Monte-Carlo Shell Model (MCSM) to tackle the high-dimensional problem is to reduce the dimensions of the most general and very large Hamiltonian by focusing on the more important parts i.e. ‘importance truncation’. The identification of these important parts is done using a Quantum Monte Carlo Diagonalization (QMCD) method [67]. The MCSM has two major characteristic features: the feasibility of including many single-particle orbits and the capability of handling many valence nucleons [67]. When this method is combined with the most recent computational advances, such as e.g. using massively parallel supercomputers [66], it allows for the calculation of a wide variety of nuclei across the nuclear chart from light to medium heavy and recently also in the lead region [1] as will be shown in the articles added to this work.

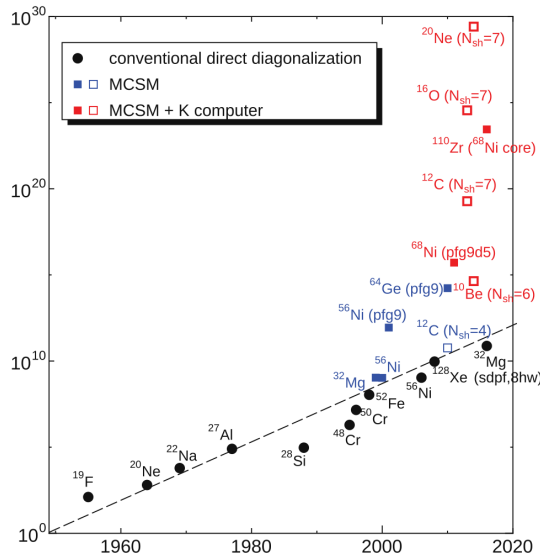


Figure 2.5: Dimension of the shell-model calculations with the Lanczos method (filled circles) and the MCSM (blue and red squares) plotted against the year of publication or presentation. The N_{sh} denotes the number of major harmonic-oscillator shells for MCSM calculations without a fixed core as basis. The dashed line is drawn to guide the eye. Figure from [66]

The MCSM method handles both odd and even-mass nuclei, and is able to study a wide variety of nuclear-structure characteristics. The strength of the MCSM lies in its possibility to calculate problems of much larger dimensions than traditional large-scale shell-model calculations, as shown in Figure 2.5. This allows for the possibility for some lighter nuclei to be computed using no inert core to start from as would normally be done for shell model calculations. These are called no-core shell-model calculations. For mercury, MCSM calculations shown in this work were performed by T. Otsuka, Y. Tsunoda and co-workers. These are the largest ever calculations of this type for such heavy nuclei and are described in the papers reported in chapter 4.

2.2.3 Deformed shell (Nilsson) model

In section 2.2.1, the assumption of a spherically symmetric nuclear potential was implicit. By relaxing this constraint on the shape of the nuclear potential, it is possible to describe the motion of single particles in a deformed nuclear potential depending on the amount of deformation. Nilsson performed calculations for non-spherical nuclear potentials around 1955 [68]. The deformed shell model, or Nilsson model, has been very successful in describing features of single-particle levels in hundreds of deformed nuclei [59].

The most common form of deformation is quadrupole deformation. Nuclear shapes with quadrupole-deformation parameters β_2 such that $\beta_2 = 0$, $\beta_2 < 0$ and $\beta_2 > 0$ are referred to as spherical, oblate and prolate shapes respectively as shown in Figure 2.6.

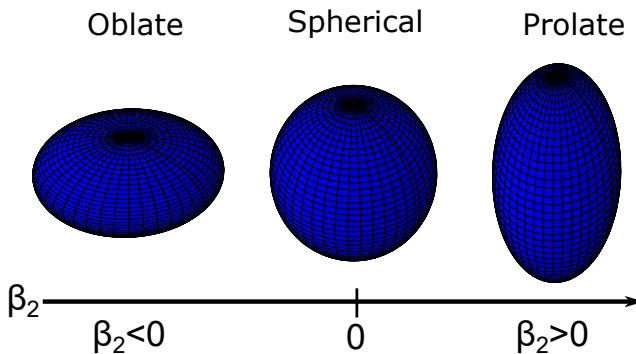


Figure 2.6: Nuclear shapes corresponding to a spherical, oblate and prolate shape with $\beta_2 = 0$, $\beta_2 < 0$ and $\beta_2 > 0$.

The deformation parameter β_2 , refers to the parametrization of a nuclear surface $R(\Theta, \phi)$ as an expansion of spherical harmonics $Y_{\lambda\mu}$ and multipole λ deformation coordinates $\alpha_{\lambda\mu}$ as

$$R(\Theta, \phi) = R_0 \left(1 + \alpha_{00} + \sum_{\lambda=1}^{\infty} \sum_{\mu=-\lambda}^{\lambda} \alpha_{\lambda\mu}^* Y_{\lambda\mu}(\theta\phi) \right) \quad (2.5)$$

where R_0 is the radius of a sphere with the same volume [46]. When axial symmetry is assumed, all $\alpha_{\lambda\mu}$ with $\mu \neq 0$ vanish and $\alpha_{\lambda 0}$ are usually referred to as β_λ . In the case of quadrupole deformation ($\lambda = 2$), β_2 is given as

$$\beta_2 = \frac{4}{3} \sqrt{\frac{\pi}{5}} \frac{\Delta R}{r_{\text{sph}}} \quad (2.6)$$

where ΔR is the radial difference between the axis of the ellipsoid and r_{sph} refers to the radius of a spherical nucleus within the LDM.

Solving the Schrödinger equation for a non-spherical potential removes ℓ as a good quantum number. This lifts the $(2j+1)$ degeneracy of the spherical shell-model energy levels. States can be labelled however by other quantum numbers e.g. the projection of j on the symmetry axis of the nuclear core which is usually denoted as Ω (or also K). Reflection symmetry of the nucleus preserves a 2-fold symmetry as $\pm\Omega$ states have the same energy. An example of a Nilsson diagram is given in Fig. 2.7, where the single particle energies are given in units of oscillator frequency $\hbar\omega_0 = 41 A^{-1/3}$ MeV, which are plotted with respect to ε_2 . The parameter ε_2 is related to the β_2 deformation parameter according to

$$\beta_2 = \sqrt{\frac{\pi}{5}} \left(\frac{4}{3} \varepsilon_2 + \frac{4}{9} \varepsilon_2^2 + \frac{4}{27} \varepsilon_2^3 + \dots \right) \quad (2.7)$$

and is to a good approximation nearly the same as β_2 . In the Nilsson diagram shown in Figure 2.7, the states are labeled as $\Omega^\pi [N n_z \Lambda]$, where N corresponds to the total oscillator shell quantum number; n_z to the number of oscillator quanta in the z direction; Ω to the projection of total angular momentum j (sum of orbital ℓ and spin s) of the odd nucleon on the symmetry axis; and Λ , the projection of angular momentum along the symmetry axis where $\Omega = \Lambda + \Sigma = \Lambda \pm 1/2$ and Σ is the projection of intrinsic spin along the symmetry axis [69].

2.3 (Beyond-)Mean Field models

In mean-field methods, the two-body nucleon-nucleon force acts as a starting point and is used to obtain optimized single-particle states in a self-consistent way, using Hartree-Fock (HF) theory [8]. Mean field wave functions are usually constructed using a HF method with pairing correlations added. The nuclear density is constrained to exhibit different deformations. As a result of these calculations, energy surfaces with different topologies for different nuclei are produced. The minimum of the energy-surface is then often found for a non-spherical density distribution: a deformed nucleus. One of the main advantages of the mean-field approach is that it can be applied to nuclei across the whole nuclear chart from light to superheavy nuclei [70].

Energy surfaces of mean-field calculations are often shown on so-called (β, γ) -planes as shown in Figure 2.8.¹ Here, the results of [63] for the even-mass mercury isotopes near the neutron $N = 104$ mid-shell are depicted.

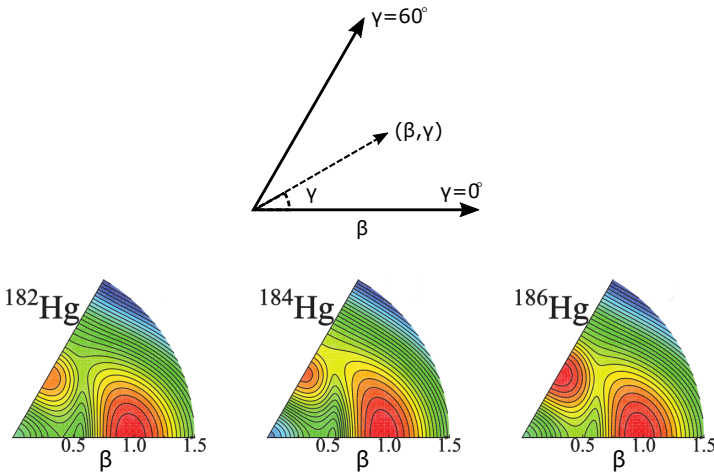


Figure 2.8: (top) schematic representation of a $\beta - \gamma$ plane. (bottom) Energy surface for IBM-CM calculations for $^{182-186}\text{Hg}$ (see section 2.2.2). The energy spacing between adjacent contour lines equals 100 keV and the deepest energy minimum is set to zero, corresponding to the red color. Figure from [63].

¹The Hill-Wheeler coefficients (β, γ) are used to describe the nuclear deformation [71]. They are related to $\alpha_{\lambda\mu}$ according to $\alpha_{20} = \beta_2 \cos \gamma$ and $\alpha_{22} = \frac{1}{\sqrt{2}} \beta_2 \sin \gamma$. When $\gamma = 0^\circ(60^\circ)$, the nucleus has a prolate (oblate) shape, while β_2 refers to the degree of deformation. If $0^\circ < \gamma < 60^\circ$, the nucleus is triaxially shaped.

In mean-field models, the motion of nucleons as independent particles is derived starting from effective forces. These effective interactions are tuned to their primary use in mean-field calculations. The form of the effective energy functional is motivated from *ab initio* theory, but the actual parameters are adjusted by global fits to experimental nuclear-structure data (e.g. masses, charge radii). A few examples of interactions that are in use today include Gogny, Skyrme or UNEDF. While at first, these calculations were only valid for ground-states properties of even nuclei, developments opened up the possibility to describe other observables such as e.g. rotational spectra of (super)heavy nuclei [45]. It is important that all symmetries that were broken in the HF approach are restored to construct states with good proton and neutron numbers, isospin and angular momentum. These states then form the basis to determine collective dynamics (mixing, energy spectra) which is referred to as the beyond-mean field approach [8].

For mercury in specific, beyond mean-field calculations were recently performed by Yao and co-workers [65] together with a study of neighbouring even- Z elements in an attempt to describe the multitude of data gathered by different experiments. As in the IBM shell-model calculations of [63], they are restricted to the even-mass mercury isotopes. Yao et al. started with mean-field wave functions with a constraint on the axial quadrupole moment and made use of the Skyrme energy density functional. To go beyond the mean field, projections on angular momentum and particle number were performed before mixing the resulting wave functions. They note an energy-surface precision better than 100 keV due to a very fine discrete Cartesian mesh. Results showed a qualitative agreement with data, but the authors concluded that the energy-density functionals have to be improved on order to achieve better results quantitatively.

Very recently, density functional theory (DFT) calculations were performed for the mercury isotopes by W. Reyssens, P-H. Heenen, M. Bender and by J. Dobaczewski and A. Pastore. Both groups described independently a different origin that would be able to account for experimentally-observed shape-staggering detailed in this thesis. The first group made use of a Skyrme-type functional, while the second one used the UNEDF interaction. The results of both groups were added to the theoretical part of Article II that can be found in chapter 4.

Chapter 3

Theoretical basics for laser spectroscopy

In laser-spectroscopy experiments for nuclear-physics research, lasers are used to probe the atomic structure to access, in a nuclear-model-independent way, nuclear properties such as the nuclear spins, magnetic dipole moments, electric quadrupole moments, and charge radii of (exotic) isotopes in their ground and isomeric states [72]. This information is found from measurements of the isotope shift (IS) and hyperfine structure (HFS) of an atomic transition. By measuring, with high precision, the resonance frequencies of electronic transitions between different hyperfine levels of an atom, the IS and HFS can be obtained. In this chapter, the theoretical basics are discussed that are needed for extracting nuclear information from experimental spectra.

During the last decades of optical spectroscopy research, several review and data evaluation papers on nuclear moments and charge radii measurements have been published. A few excellent ones can be found in [17, 21, 53, 72–76].

3.1 Hyperfine structure and nuclear moments

The so-called hyperfine structure of electronic orbitals arises from the coupling of the electronic and nuclear angular momenta I and J [53]. This coupling lifts the degeneracy of the atomic fine-structure energy levels, when both I and J are larger than zero, into the hyperfine levels labelled by the total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$ of the combined nucleus and electron system. The amount of levels into which the degeneracy gets lifted is given by $|I - J| \leq F \leq |I + J|$,

where F increments by unit values. As an example, under the assumption that ^{179}Hg has a ground state with nuclear spin $I^\pi = 7/2^+$, the $6s6p^3P_1$ orbital with $J = 1$ is split into three hyperfine levels with F -values ranging from $5/2$ to $9/2$. No degeneracy is lifted for the $6s^2\ ^1S_0$ due to the $J = 0$ nature of this excited state. Figure 3.1 shows the transition with $6s^2\ ^1S_0$ as lower level and $6s6p^3P_1$ as upper level being split into their hyperfine components.

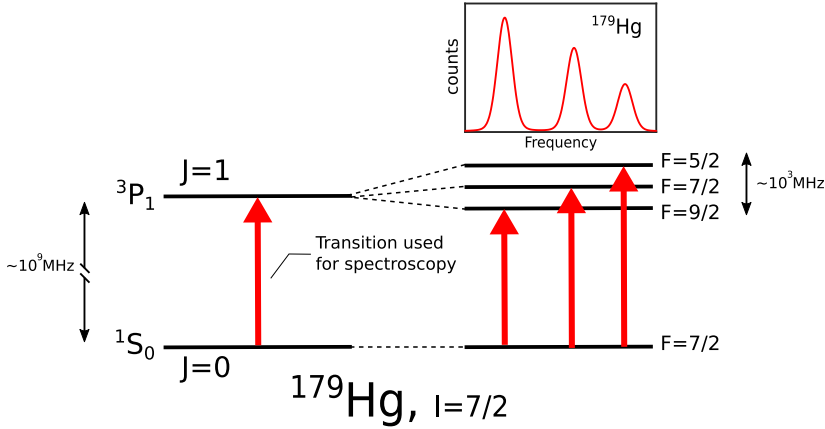


Figure 3.1: Hyperfine level structure for ^{179}Hg of the $6s^2\ ^1S_0 \rightarrow 6s6p^3P_1$ transition. The right-hand side shows the splitting of the 3P_1 level into different F -states for an $I = 7/2^-$ nuclear spin. An exemplary spectrum is given on top where the resonance peaks correspond to the transitions shown by the red arrows below.

The energy with which the respective components are split is dictated by the hyperfine parameters a and b , which are related to the magnetic dipole and electric quadrupole moment of the nucleus respectively. Including only the leading terms arising from the nuclear dipole and quadrupole moments, the energy shift of a hyperfine level with respect to the fine structure level is given by equation 3.1.

$$\Delta E = \Delta E_{\text{dipole}} + \Delta E_{\text{quadrupole}} \quad (3.1)$$

$$\frac{\Delta E}{h} = a \frac{K}{2} + b \frac{0.75K(K+1) - I(I+1)J(J+1)}{2IJ(2I-1)(2J-1)} \quad (3.2)$$

where $K = F(F+1) - I(I+1) - J(J+1)$. a and b are the magnetic dipole and electric quadrupole hyperfine coupling constants respectively and are given

by the expressions

$$a = \frac{\mu_I B_e(0)}{IJ} \quad (3.3)$$

$$b = eQ_s \left\langle \frac{\partial^2 V_e}{\partial z^2} \right\rangle \quad (3.4)$$

with μ_I denoting the magnetic dipole moment of the nucleus, $B_e(0)$ the magnetic field created by the electrons at the nucleus, Q_s the spectroscopic quadrupole moment and $\left\langle \frac{\partial^2 V_e}{\partial z^2} \right\rangle$ corresponding to the electric field gradient produced by the electrons. More details can be found in [53, 74].

3.1.1 Magnetic dipole moment

The magnetic dipole moment of a nucleus with nuclear spin I larger than 0, can be evaluated from the a -parameter obtained from a fit of the hyperfine spectrum. When the magnetic moment is known for a reference isotope, the ratio of a -parameters can be used to determine the magnetic moment of the isotope under study via

$$\frac{a}{a_{\text{ref}}} \approx \frac{\mu}{\mu_{\text{ref}}} \frac{I_{\text{ref}}}{I} \quad (3.5)$$

The magnetic moment of the reference isotope should then be already measured in an independent experiment using e.g. Nuclear Magnetic Resonance (NMR) [17]. This evaluation does not take into account the hyperfine anomaly related to the change in the nuclear charge and magnetization distribution from one isotope to the other, which is explained in more detail in section 3.1.3. Therefore, eq. 3.5 is not described as exact. Often however, the hyperfine anomaly is very small and the approximation in eq. 3.5 is neglected and the formula is treated as being exact.

3.1.2 Electric quadrupole moment

Analogous to the determination of the magnetic moment, the quadrupole moment can be determined using the b -parameter extracted from the hyperfine spectrum when prior knowledge of the quadrupole moment for a reference is present.

$$\frac{b}{b_{\text{ref}}} = \frac{Q_s}{Q_s^{\text{ref}}} \quad (3.6)$$

The spectroscopic quadrupole moment, Q_s can only be determined for nuclei and atomic orbitals with spin $I, J > 1/2$. It is a sensitive probe to the nature

of deformation in atomic nuclei and can be related to the intrinsic quadrupole moment Q_0 , assuming axial symmetry, through

$$Q_s = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} Q_0 \quad (3.7)$$

Where K denotes the spin-projection on the symmetry axis of the nucleus which is presumed to have axial symmetry. This intrinsic quadrupole moment is related to the quadrupole deformation parameter $\langle\beta_2\rangle$ according to [74]

$$Q_0 \approx \frac{3}{\sqrt{5\pi}} Z \langle r^2 \rangle_{\text{sph}} \langle\beta_2\rangle (1 + 0.36 \langle\beta_2\rangle) \quad (3.8)$$

The second-order term shown is often neglected, as for instance in [53].

3.1.3 Hyperfine anomaly

For magnetic moments, the deviation from a point-like nucleus and the finite change in the distribution of nuclear charge and magnetization from one isotope to the other is the reason why equation 3.5 is an approximation. This change from one isotope to the other is referred to as the hyperfine anomaly (HFA) and arises from two effects called the “Breit-Rosenthal” (BR) [77] and “Bohr-Weisskopf” (BW) [78] effects. These effects take into account the finite spatial distribution of the nuclear charge and the distribution of nuclear magnetism within the nucleus respectively [17]. To investigate the importance of this effect, the following approach is used in [74]. If the magnetic hyperfine constant for the point-like nucleus is denoted as a_{point} , then the observable magnetic hyperfine constant a can be presented as follows:

$$a = a_{\text{point}}(1 + \epsilon)(1 + \delta) \quad (3.9)$$

where ϵ and δ refer to the BW and BR effects respectively. The ratio of magnetic hyperfine coupling constants becomes:

$$\frac{a}{a'} = \frac{a_{\text{point}}(1 + \epsilon)(1 + \delta)}{a'_{\text{point}}(1 + \epsilon')(1 + \delta')} \approx \frac{\mu I'}{\mu' I} \left(1 + {}^A\Delta^{A'}\right) \quad (3.10)$$

with

$${}^A\Delta^{A'} = {}^A\Delta_{\text{BW}}^{A'} + {}^A\Delta_{\text{BR}}^{A'} = (\epsilon - \epsilon') + (\delta - \delta'). \quad (3.11)$$

The hyperfine anomaly can be determined if independent values for magnetic moment μ and a -factor for the pair of isotopes under study are measured with high accuracy. A recent review of experimentally determined hyperfine anomalies is given in [79]. Usually, the anomaly is a small correction on the

percent level, but is observed in specific cases to lead to values up to 10% [17]. Direct measurements of transition energies within a hyperfine multiplet can be performed using radio-frequency resonance spectroscopy. Combined with knowledge of the nuclear g -factor, readily provides high accuracy measurements of $^A\Delta^{A'}$ [17, 79]. For mercury, the effect is estimated in chapter 4.

3.2 Isotope shift and change in mean-square charge radius

The energy of the same electronic transition i differs from one isotope or isomer to the other. This shift is referred to as the isotope or isomer shift (IS). The origin of the shift lies in the mass and size differences between different isotopes of the same isotopic chain. The IS between two isotopes with mass numbers A and A' is defined as the change in the frequency of atomic transition, $\delta\nu_i^{A,A'}$, and is usually split in two components: the Mass and Field Shifts.

$$\delta\nu_i^{A,A'} = \nu_i^{A'} - \nu_i^A = \delta\nu_{i,\text{Mass}}^{A,A'} + \delta\nu_{i,\text{Field}}^{A,A'} \quad (3.12)$$

Both contributions are explained in more detail in the following sections 3.2.1 and 3.2.2.

3.2.1 Mass Shift

The mass shift, originating from the change in mass between different isotopes can be separated into two terms: the Normal Mass Shift (NMS) and the Specific mass shift (SMS). These terms are related to the difference in reduced mass and the correlation of electronic momenta in a multi-electron system respectively [17]. The separation of the Mass Shift into the NMS and SMS can be written as

$$\delta\nu_{i,\text{Mass}}^{A,A'} = \delta\nu_{i,\text{NMS}}^{A,A'} + \delta\nu_{i,\text{SMS}}^{A,A'} = (K_{\text{NMS}} + K_{\text{SMS}}) \frac{m^A - m^{A'}}{m^A m^{A'}} \quad (3.13)$$

where $(K_{\text{NMS}} + K_{\text{SMS}})$ is often combined to a single M -factor. The Normal Mass Shift is given as

$$\delta\nu_{i,\text{NMS}}^{A,A'} = \frac{m_e}{m_p} \frac{m^A - m^{A'}}{m^A m^{A'}} \nu_i^A \quad (3.14)$$

where m_e and m_p denote the electron and proton mass respectively and ν_i^A refers to the atomic transition frequency. While the Normal Mass Shift is easily evaluated with equation 3.14, the Specific Mass Shift has to be provided from theory or be evaluated using a King plot (section 3.2.3).

3.2.2 Field Shift

The Field Shift arises from the difference in the Coulomb potential between isotopes probed by the electronic wave functions at the nucleus. The Field Shift is given by

$$\delta\nu_{i,\text{Field}}^{A,A'} = \frac{Ze^2}{6h\epsilon_0} \Delta|\Psi(0)|^2 \delta\langle r^2 \rangle^{A,A'} \quad (3.15)$$

and where $\Delta|\Psi(0)|^2$ describes difference in overlap of the electronic wavefunctions of the atomic levels at the nucleus, and $\delta\langle r^2 \rangle^{A,A'}$ denotes the difference in mean-square charge radius between the isotope pair. More often, the Field Shift is reformulated as

$$\delta\nu_{i,\text{Field}}^{A,A'} = F(Z)\lambda^{A,A'} \quad (3.16)$$

where $F(Z)$, the electronic F -factor, contains the information on the electronic configuration and $\lambda^{A,A'}$ is in fact a series expansion containing all higher order mean charge radii [80]. The contribution of these higher order moments is however usually taken into a single k -factor, such that the expression for $\lambda^{A,A'}$ becomes $\lambda^{A,A'} = k \delta\langle r^2 \rangle^{A,A'}$. A combination of the Field and Mass Shifts resulting in the total isotope shift is given by equation 3.18.

3.2.3 King plot

In order to extract information on the difference in mean-square charge radius from the measured IS, the F and M -factors of the total isotope shift in equation 3.18 have to be known. These two factors can either be provided by non-trivial atomic-theory calculations or by a King-plot analysis using only experimental data.

A King plot combines isotope-shift measurements for isotopes measured by two different atomic transitions from which information on the electronic F and M factors can be obtained. The linear relation that exists between the modified isotope shifts $(\delta\nu_{i,\text{Field}}^{A,A'} \cdot \frac{A-A'}{A^*A'})$ has a slope κ that equals the F -factor ratio ($\kappa = F_i/F_{i_0}$) of the two transitions i and i_0 . Information on the M factors can be found in the intercept s via $s = M_{i_0} + \kappa M_i$.

3.2.4 Mean-square charge radius

The nuclear mean-square charge radius $\langle r^2 \rangle$ is defined as

$$\langle r^2 \rangle = \frac{\int_0^\infty \rho(\vec{r}) r^2 dr^3}{\int_0^\infty \rho(\vec{r}) dr^3} \quad (3.17)$$

where $\rho(\vec{r})$ describes the charge distribution of the nucleons in the nucleus. The difference in mean-square charge radius is extracted from the IS according to equation

$$\delta\nu_i^{A,A'} = M \frac{m^A - m^{A'}}{m^A m^{A'}} + F(Z) \lambda^{A,A'} \quad (3.18)$$

$$= (K_{NMS} + K_{SMS}) \frac{m^A - m^{A'}}{m^A m^{A'}} + F(Z) k \delta \langle r^2 \rangle^{A,A'} \quad (3.19)$$

The mean-square charge radius is related to the deformation of the nucleus according to

$$\langle r^2 \rangle = \langle r^2 \rangle_{\text{DM}} \left(1 + \frac{5}{4\pi} \langle \beta_2^2 \rangle \right) \quad (3.20)$$

with $\langle r^2 \rangle_{\text{DM}}$ denoting droplet-model prediction for the mean-square charge radius of a spherical nucleus of identical volume and β_2 is the second-order nuclear deformation parameter (see section 2.2.3). This relation is what makes the measurement of the isotopic shift and resulting evaluation of the difference in mean-square charge radius of an isotopic pair such a sensitive probe for their difference in nuclear deformation [17].

Two different observables can be linked to $\langle \beta_2^2 \rangle$. While the charge radius is related to $\langle \beta_2^2 \rangle$, the sign and magnitude of static deformation can be directly obtained from the hyperfine structure (see sec 3.1.2) and together, they can provide an indication of the nature of the nuclear deformation [81]. If $\langle \beta_2^2 \rangle$ obtained from charge radii measurements is much larger than $\langle \beta_2 \rangle^2$ from the hyperfine structure, a sizeable contribution to $\langle \beta_2^2 \rangle$ might be coming from dynamical deformation according to [53]

$$\underbrace{\langle \beta_2^2 \rangle}_{\substack{\text{total} \\ \text{(charge radius)}}} = \underbrace{\langle \beta_2 \rangle^2}_{\substack{\text{static} \\ \text{(hfs)}}} + \underbrace{\langle \beta_2^2 \rangle - \langle \beta_2 \rangle^2}_{\text{dynamic}} \quad (3.21)$$

3.3 Broadening mechanisms of spectral lines

The observed hyperfine spectrum of an isotope is subject to a convolution of line broadening mechanisms and shifts. These mechanisms obscure the nuclear information present in the obtained spectrum. The dominant type of broadening present in a spectrum depends on the experimental technique and the laser system used as well as on the isotopic ensemble conditions. Here, the main sources of line-shape broadening are discussed without giving a rigorous derivation. More information on the mathematics and quantum optics involved in these processes can be found in [82, 83].

3.3.1 Natural line width

As a consequence of the Heisenberg uncertainty principle, $\Delta t \Delta E \approx h$, the energy of an excited state can never be known exactly due to the finite lifetime of this state. In laser spectroscopy experiments, this effectively leads to a Lorentz-type broadening of the resonance lines, where the function $f(\nu)$ that can be used to describe the spectral line shape is of the form

$$f(\nu) = \frac{\Gamma_{\text{nat}}}{4\pi^2} \frac{1}{(\nu - \nu_0)^2 + (\Gamma_{\text{nat}}/4\pi)^2} \quad (3.22)$$

where Γ_{nat} is the Lorentz width related to the spontaneous decay rate of the probed atomic state. Most often, the natural line width is not the main contributor to the line profile of a spectrum, but rather poses a lower limit. The 3P_1 level used in the laser-spectroscopy experiment on mercury discussed in this work, decays exclusively to the ground state via a transition which has a strength of $A = 8.4 \times 10^6 \text{s}^{-1}$ [84]. This corresponds to a natural line width of 1.3 MHz, which is negligible in comparison to other contributions.

3.3.2 Doppler broadening and shift

Atoms in an ensemble which have differing relative velocities with respect to the laser light, perceive a Doppler-shifted frequency of the laser light according to the formula

$$\Delta\nu = \nu_0 \frac{v_r}{c} \quad (3.23)$$

where v_r corresponds to the atoms velocity component parallel to the incoming laser light. When the atomic ensemble has a net velocity difference with respect to the lab frame, one has to correct for the Doppler shift. A distribution of relative atomic velocities parallel to the laser light thus causes a broadening of

the line profile when measuring e.g. a hyperfine spectrum. Thermal motion in an atomic ensemble therefor results in Doppler broadening. This effectively broadens the line shape according to a Gaussian spectral shape $f(\nu)$ depending on the temperature T as

$$f(\nu) = I_0 \exp \left[- \left(\frac{c(\nu - \nu_0)}{\nu_0 v_p} \right)^2 \right] \quad \text{where} \quad (3.24)$$

$$v_p = (2KT/m)^{1/2} \quad (3.25)$$

$$\text{FWHM}(f) = \frac{\nu_0}{c} \sqrt{\frac{8KT \ln(2)}{m} + v_{\text{turb}}^2} \quad (3.26)$$

with $\text{FWHM}(f)$ corresponding to the effective Doppler width [82]. In equations 3.24-3.26, I_0 refers to the Gaussian amplitude, while v_p corresponds to the most probable velocity according to the Maxwell-Boltzmann distribution at temperature T with K corresponding to the Boltzmann constant. For in-source resonance ionization spectroscopy, it is the Doppler contribution to the total spectral line width that is dominant. Excluding turbulent motion, the Doppler width, corresponding to half of the induced full width at half maximum (FWHM) of equation 3.26, for the 253 nm transition in mercury corresponds to 2.7 GHz at a temperature T of 2000 K.

3.3.3 Pressure broadening and shift

Pressure broadening and shift are effects on the line shape due to atomic collision. The amount of broadening and shift in a gaseous environment depends on the electronic configuration of both collisional partners as well as on the pressure. Usually the broadening takes on a Lorentzian line profile, but the effect can also induce line-shape asymmetries [82]. Typical broadening widths are of the order of ≈ 10 MHz/mbar, while pressure shifts are of the order of a few MHz/mbar [13]. Other than just broadening and shifting the line shape, the interaction between an isotope and residual gas atoms could result in molecule formation, charge exchange or induced decay towards an atomic state that might not be probed in the experiment and thus an overall loss in signal. In an in-gas-cell resonance ionization experiment, the pressure broadening becomes the main contributor to the overall broadening at pressures $\gtrsim 100$ mbar [85].

3.3.4 Power broadening

Pressure or collisional broadening originates from the laser-power induced increase of the decay rate of an atomic level. Due to the homogeneous nature of this effect (i.e. all atoms are affected in the same way), this broadening adopts a Lorentzian profile. Power broadening provides a qualitatively similar result as pressure or collisional broadening, the difference here being that the increased decay rate originates from strong laser fields rather than atomic collisions [83]. In order to ensure that in experiment, power broadening does not affect the precision with which the nuclear information can be extracted, it is often advised to stay within the linear regime of a so-called saturation curve, which relates the signal strength to the laser power. In resonance-ionization spectroscopy, both power-density effects as well as temporal overlap of both the excitation and ionization laser affect the overall line width [86]. Both parameters can usually be easily adjusted to obtain the preferred efficiency to precision ratio.

Chapter 4

Laser spectroscopy of mercury isotopes

The majority of relevant information related to the laser-spectroscopy experiment of the mercury isotopes performed at ISOLDE, CERN has resulted in the preparation of two papers (**Articles I** and **II**) reproduced in section 4.4. In this chapter, more extended information is given in addition to both articles.

4.1 Experimental setup at ISOLDE, CERN

The experiment took place at the Isotope Separator On-Line DEvice (ISOLDE) in CERN, Geneva [87] (Figure 4.1). This facility is an ISOL-type radioactive ion beam facility, where 1.4 GeV protons coming from CERN's PS Booster (PSB) impinge on a thick target. The PSB delivers pulsed proton beams, whereby the bunches are structured in a super cycle (SC) consisting of $\sim 2 \mu\text{s}$ -long pulses separated by about 1.2 s. Each pulse can contain up to 3×10^{13} protons. For this experiment, the target-ion source combination consisted of a molten-lead target, coupled to a Variable Arc Discharge Laser Ion Source (VADLIS) type ion source. The latter is an extended version of a typical Variable Arc Discharge Ion Source (VADIS), that can be operated in VADIS, VADLIS or Resonance Ionization Laser Ion Source (RILIS) mode, depending on the experiment's yield and contamination requirements [88]. In this specific experiment, the ion source was run in RILIS mode, where the non-resonant 'discharge'-type ionization is reduced by decreasing the energy of the electrons below the electron-impact ionization threshold, and resonant laser ionization is the main process

of ionization. Using a non-resonant ionization mode, would add to the beam contamination which might add constant background to the resulting hyperfine scans.

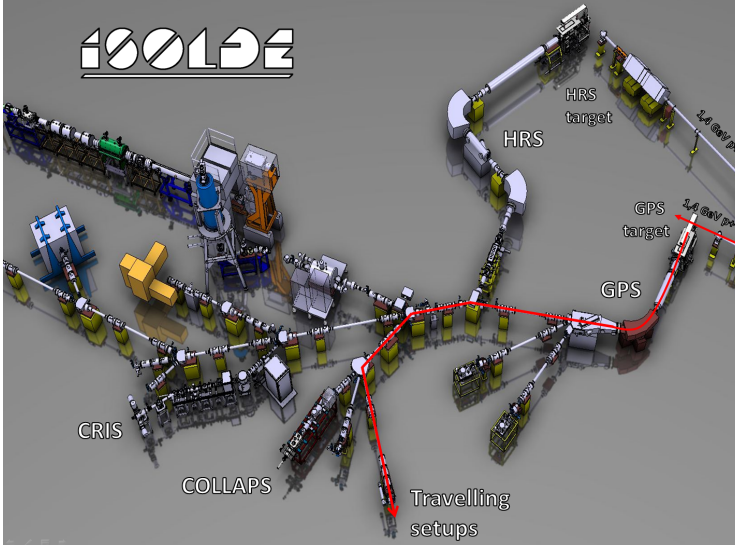


Figure 4.1: ISOLDE layout. Picture adjusted from [89]

Reaction products effused out of the heated target kept at high temperature to enhance effusive properties, through a heated spiral chimney into the ion-source cavity, where resonant laser ionization took place using the RILIS lasers (sec 4.1.1). The target and ion-source were all kept at high voltage of +30kV relative to ground potential. The resonantly produced ions were extracted by a grounded extraction electrode. They were subsequently mass-separated by the General Purpose Mass separator (GPS), before being sent to one of three detection setups: the ISOLDE Faraday Cup (FC), the Windmill (WM) and ISOLTRAP's Multi-reflection Time-of-flight mass separator/spectrometer (MR-TOF-MS). The path of the protons and of the mercury ions is shown on Figure 4.1 for the case when Windmill detection setup was used. A schematic view of the experimental setup, including the three different detection setups and the RILIS lasers, is shown in Figure 4.2.

The choice for different detection techniques was based on several factors such as beam intensity, isotope half-life, production yield, decay mode and branching ratio. The Faraday Cup was used in cases where the ion-beam current intensity was in the order of pA . The Windmill system was used for alpha-decaying

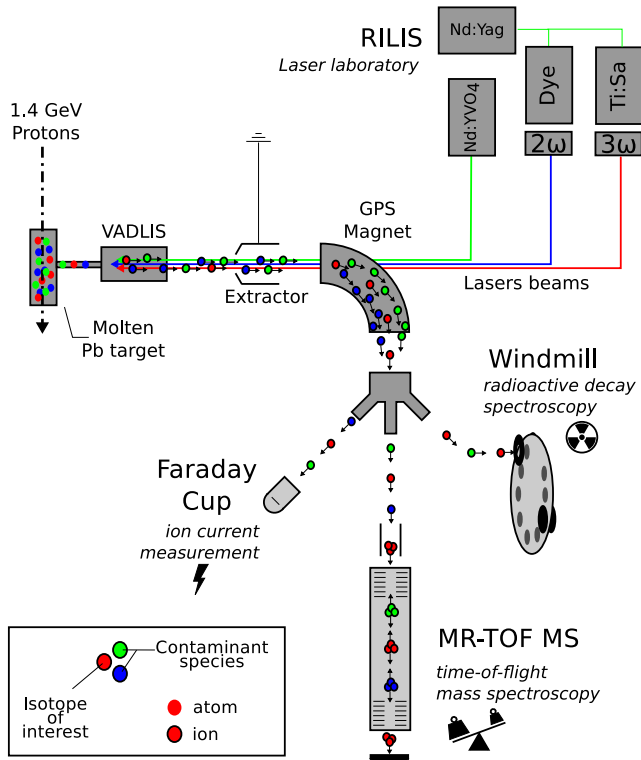


Figure 4.2: Schematic overview of the experimental setup.

isotopes with low production yields ($\sim 3 \times 10^{-2} \alpha's/s$ for ^{177}Hg) with a short half-life. Longer-lived contamination might build up in the carbon foils adding to an enlarged background, and β -decaying isotopes only provide a mono-energetic fingerprint through subsequent γ -decay that is detected with lower efficiency. Therefore, the MR-TOF technique was used in cases where β -decay was the main form of decay and where the beam intensity was too small to be measured by the Faraday Cup.

Having a resolving power ($R = M/\Delta M$) of 1000, the GPS magnet should be able to separate neighboring mercury isotopes with masses of for instance $A=177$ and 178 . However, due to an incomplete acceleration or possible collisions with rest gas in combination with the non-ideal emittance of the initial ion distribution and higher production cross sections of less-exotic neighbouring isotopes, beam contamination is still present. This is clearly shown in Fig. 4.11 of section 4.2, where contamination up until $A=183$, could be detected, even

when the central mass of the magnet was set at $A = 177$.

The use of a molten lead target was decided on after analyzing the results of a preparatory experiment, where the yield of different isotopic species of mercury were compared for different target-ion source combinations. The measurement times and corresponding maximum event rates on resonance with the RILIS lasers in narrowband mode are shown in table 4.1. For data obtained with the WM setup, the summed count rate of both implantation detectors is given in α 's/s. The count rate measured with the MR-ToF setup is given in counts/shot, where each shot refers to a single bunch of accumulated ions ejected from the buncher (see section 4.1.3). The measurement time indicated in table 4.1 for the WM measurements refers to the total time for all files obtained for a certain isotope given by a 100 Hz pulser. For MR-ToF measurements, this time refers to the total HFS-scanning time for each isotope. The yield data extracted from these count rates are summarized in Fig 3 of Article II.

Table 4.1: Setup, measurement time and corresponding maximum event rate for the isotopes of interest on resonance during HFS scans of laser-spectroscopy experiment IS598. For the windmill measurements, the obtained α rate is given for both silicon detectors at the implantation site. For MR-ToF measurements, the production rate is given in counts/shot.

Isotope	Setup	Measurement time	α rate (s^{-1})
^{177}Hg	WM	8 hr 30 min	0.03
^{178}Hg	WM	4 hr 14 min	1.0
^{179}Hg	WM	2 hr 46 min	3.8
^{180}Hg	WM	6 hr 26 min	50
^{181}Hg	WM	6 hr 9 min	179
^{182}Hg	WM	0 hr 42 min	324

Isotope	Setup	Measurement time	Ion rate (counts/shot)
^{183}Hg	MR-ToF	1 hr 6 min	11.48
^{184}Hg	MR-ToF	0 hr 35 min	19.72
^{185}Hg	MR-ToF	1 hr 38 min	28.81
^{198}Hg	MR-ToF	1 hr 37 min	15.91
^{202}Hg	MR-ToF	0 hr 30 min	23.64
^{203}Hg	MR-ToF	0 hr 56 min	25.25
^{206}Hg	MR-ToF	0 hr 31 min	14.0
^{207}Hg	MR-ToF	1 hr 32 min	15.29
^{208}Hg	MR-ToF	2 hr 15 min	1.34

4.1.1 Resonant laser ionization

In this experiment, the Resonant Ionization Laser Ion Source (RILIS) at ISOLDE made use of the ionization scheme shown in Figure 4.3. This 3-step scheme, consisting of two resonant steps and a final non-resonant ionization step, detailed in [90], has a measured total target-ion source efficiency of 6%. Spectroscopy was performed on the 253.65 nm $6s^2\ ^1S_0 \rightarrow 6s6p\ ^3P_1$ transition producing well-resolved hyperfine spectra even at high temperature. This is a usual necessity of in-source ionization, as the high temperature of the atomic ensemble dictates the Doppler broadening and spectral resolution of the hyperfine spectra.

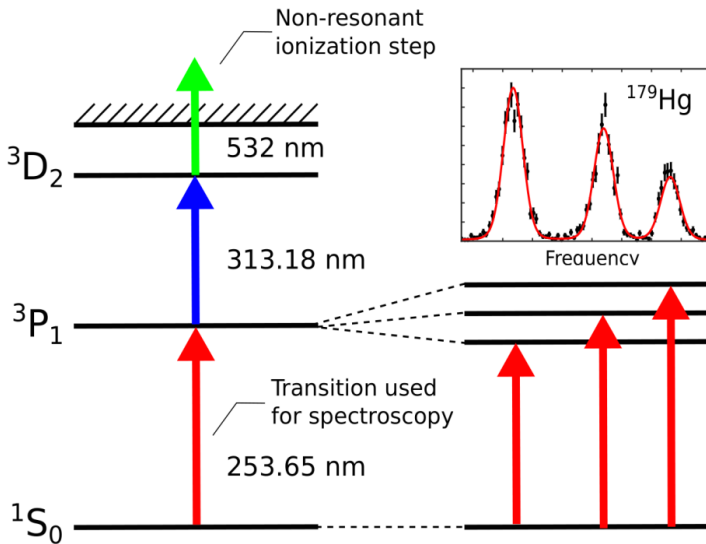


Figure 4.3: Ionization scheme of mercury that was used during the experiment and developed in [90].

The first step, a resonant step of 253.65 nm is produced by frequency-tripling the light of a Nd:Yag-pumped Ti:Sa laser with a spectral linewidth of ~ 1.5 GHz after tripling. Following the first step, was a 313.18 nm step produced by frequency-doubling the light produced by a Sirah Credo Dye laser, operated with an ethanol solution of DCM dye. Lastly, the light for the non-resonant 532 nm ionization step was produced by a Nd:YVO4 Lumera Blaze laser [90]. The laser linewidth of the first-step laser is rather broad with respect to laser systems commonly used in high-resolution laser spectroscopy experiments, which typically have spectral linewidths in the order of ~ 10 -100 MHz. However, this option is the best suitable because of the broad spectral distribution of the

high-temperature ensemble used in in-source spectroscopy. Using laser systems with smaller widths would result in a loss of efficiency as part of the ensemble would not be excited. It is therefore best practice to match the laser linewidth with the optical properties of the probed ensemble for optimal operation such that neither a loss of efficiency nor an extra induced broadening result from non-optimal spectral matching.

4.1.2 Windmill detection setup

Implantation and detector setup

The windmill detection setup (WM) [91], shown in Figure 4.4, consists of a vacuum chamber holding a rotating wheel that houses 10 thin carbon foils of 10 or 12 mm diameter ($20 \mu\text{g}/\text{cm}^2$) [92]. Surrounding these foils are two pairs of silicon detectors. The first pair is positioned around the C-foil in which the beam is implanted. This pair consists of an annular surface barrier detector (Ortec, TC-025-450-300-S, 6 mm hole, 450 mm^2 active area) through which the beam passes, and a circular surface barrier detector (Ortec, TB-020-300-500, 300 mm^2 active area) placed at either side of the wheel. After implantation of the beam for the duration of 1 to 5 super cycles, the wheel rotates and a fresh foil is placed in view of the beam. The foil that was irradiated is moved between a second pair of detectors placed at 144° from the implantation site with respect to the wheel center. Located at the ‘decay site’, are two Silicon PIPS detectors (Canberra, PD 300-15-300 RM, 300 mm^2 active area) which are used to study longer-lived radioactive isotopes that were implanted directly or those produced via the decay of previously implanted nuclei.

The total alpha particle detection efficiency at the implantation site was measured to be 34% and a detector resolution of 35 keV FWHM was obtained. Surrounding the WM vacuum chamber were two Germanium detectors used to study X-rays or gamma radiation from the implanted isotopes. These detectors can also be used to study alpha-gamma or electron-gamma coincidences. Both Ge detectors were aimed at the implantation site of the WM. The Low-Energy Ge detector (LEGe, planar Canberra detector model 771510, $400 \mu\text{m}$ thin Be window, 44 mm active diameter, 1500 mm^2 active area) was positioned in line with the ion-beam direction behind the chamber, where the aluminium back-plate thickness of the chamber is reduced down to 0.2 cm to increase low-energy gamma- or X-ray detection efficiency. A high-purity Ge detector (HPGe, model GC7520, 75% relative efficiency) was placed at a 90° angle to the beam path.

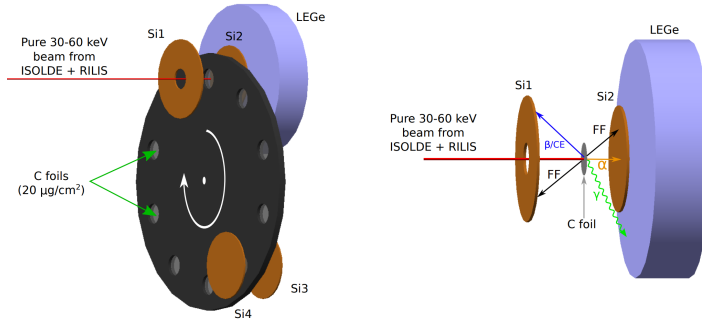


Figure 4.4: Model of the Windmill detection setup. Left: The silicon detector pairs (brown), carbon foils (light grey), rotatable wheel (black) and the Low-Energy germanium detector (LEGe, blue) are shown. The germanium detector positioned at a 90° angle with the beam path is not visible in this sketch. Right: zoomed in view of the implantation site with α particles (orange), β /conversion electrons (blue), γ rays (green) and fission fragments (FF, black) originating from the foil position. Figure from [93]

The silicon detectors were calibrated using the α -decay of the observed $^{182-178}\text{Hg}$ isotopes and their α and β -daughters $^{176-174}\text{Pt}$ and ^{181}Au . As the calibration of detector Si2 was shifting significantly over time, the calibration curve was refitted for different groups of data files. The germanium-detector energy calibrations were performed with the known γ -ray energies of ^{133}Ba (6.686 kBq) and ^{152}Eu (7.953 kBq) calibration sources. The fits were performed with a linear function. An example of a calibration for the germanium (LEGe and HPGe, referred to as Ge1 and Ge2) and silicon (Si1 and Si2) detectors is shown in Figure 4.5.

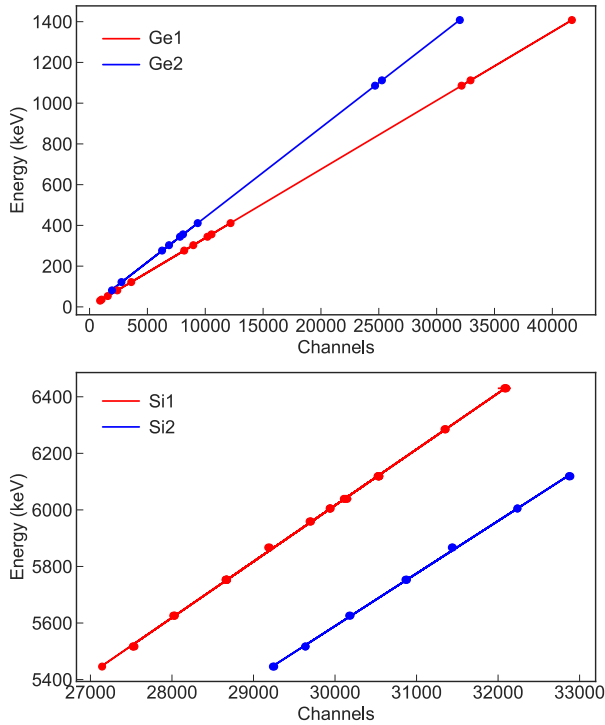


Figure 4.5: Calibration curves for germanium (top) and silicon (bottom) detectors.

Timing and DAQ

The scanning of the hyperfine spectra was automated using a combination of analog signal processing and data exchange with RILIS depicted in Figures 4.6 and 4.7.

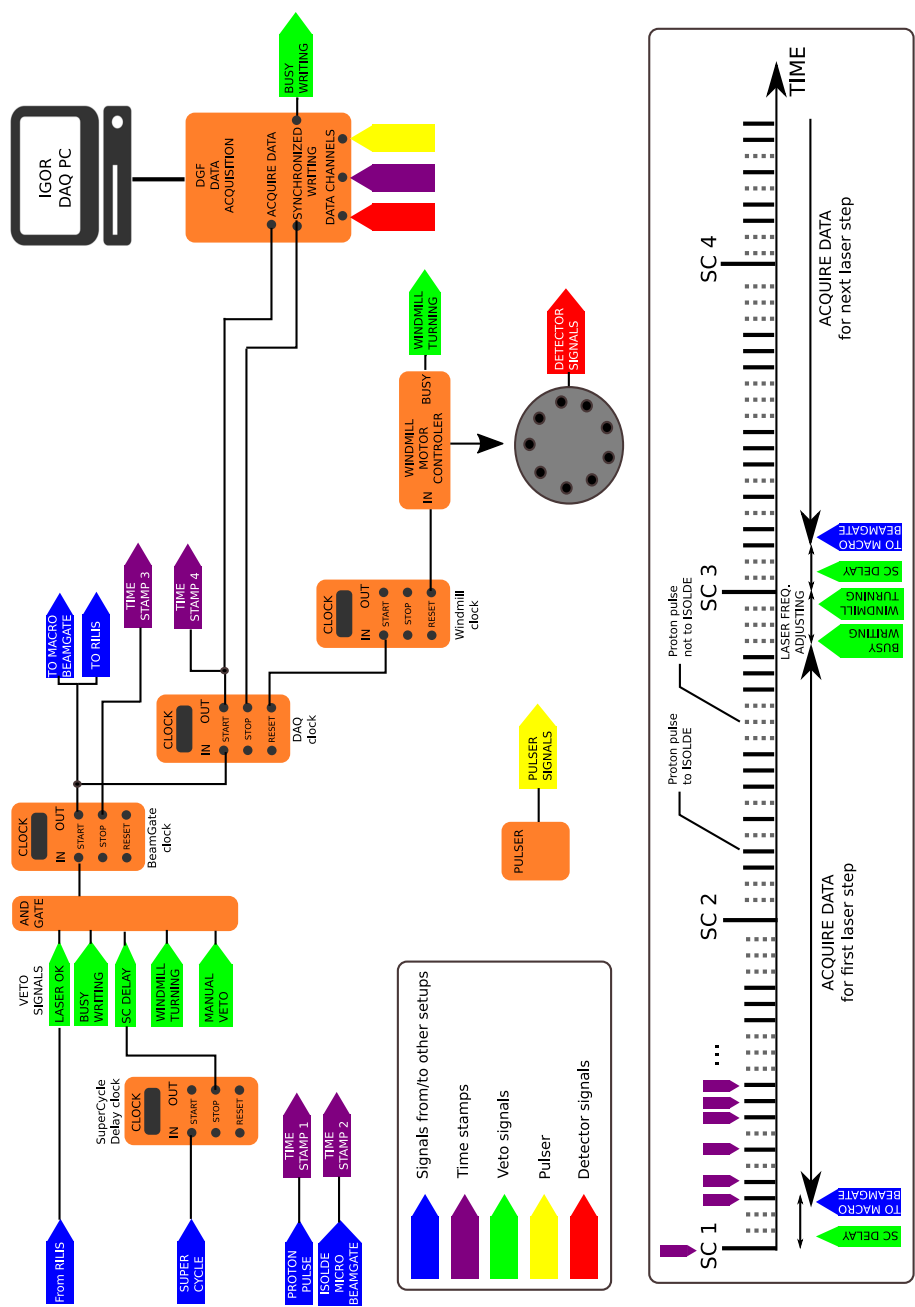


Figure 4.6: Top: Schematic data acquisition and signal processing layout. Bottom: Timeline of acquisition with several timing and veto signals superposed on the supercycle structure.

In the top of Figure 4.6, a simplified layout is shown of how the data acquisition was run, after the initial clock timings were set at the beginning of a scan. At the bottom of Figure 4.6, a typical time profile of the supercycle structure is shown, where some of the signals, referring to the top figure are indicated. Most of the setup was remotely controlled from the DAQ-room outside of the ISOLDE hall, as shown in Figure 4.7, where the computer layout and data transfer are shown. The current setup, allowed for a ‘live view’ of the hyperfine spectrum being taken both in the DAQ room and by the RILIS team, which made it possible to intervene during scans if necessary.

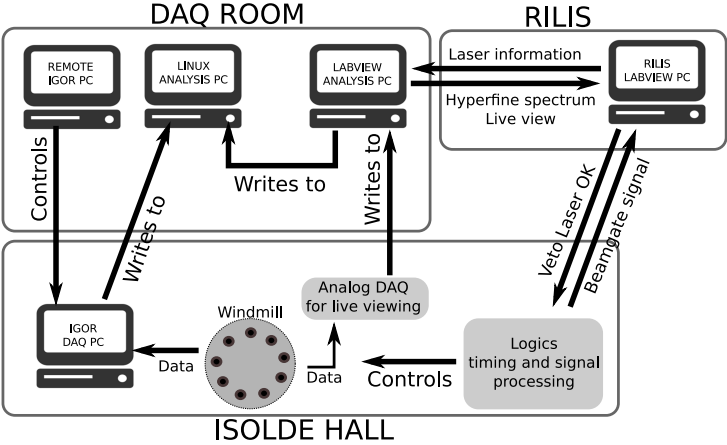


Figure 4.7: Layout of computers and information flow of the experiment

4.1.3 MR-TOF-MS

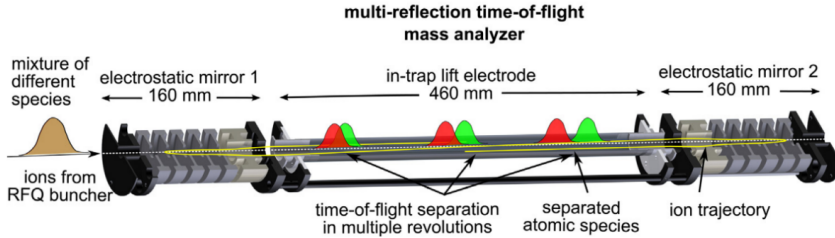


Figure 4.8: ISOLTRAP’s Multi-reflection Time-of-flight (MR-TOF) Mass spectrometer/seperator. Picture taken from [94].

ISOLTRAP’s multi-reflection time-of-flight mass separator/spectrometer device (MR-TOF MS) was used for mass separation and ion detection during laser scans of $183\text{--}185, 198, 202, 203, 206\text{--}208\text{Hg}$. This device, discussed in [94] and shown in Figure 4.8, consists of two 160 mm-long electrostatic mirrors surrounded by shielding electrodes. Ion bunches, consisting of different isotopic species from a helium-filled RFQ buncher are injected to the device, where multiple round-trips causes a mass- and charge-state dependent separation in time for different isotopic and isomeric species in each bunch. After usually ~ 1000 round-trips, ejected ions were measured on a MagnetTOFTM TOF detector, positioned behind the device.

An example of a TOF-spectrum obtained for ^{208}Hg is shown at the bottom of Figure 4.9, where the presence of isobaric lead contamination is more abundant than the resonantly ionized mercury. The time difference however, allows for a clear identification and quasi background free hyperfine spectra. At the top of Figure 4.9, all MR-TOF flight times and count rates per shot are shown.

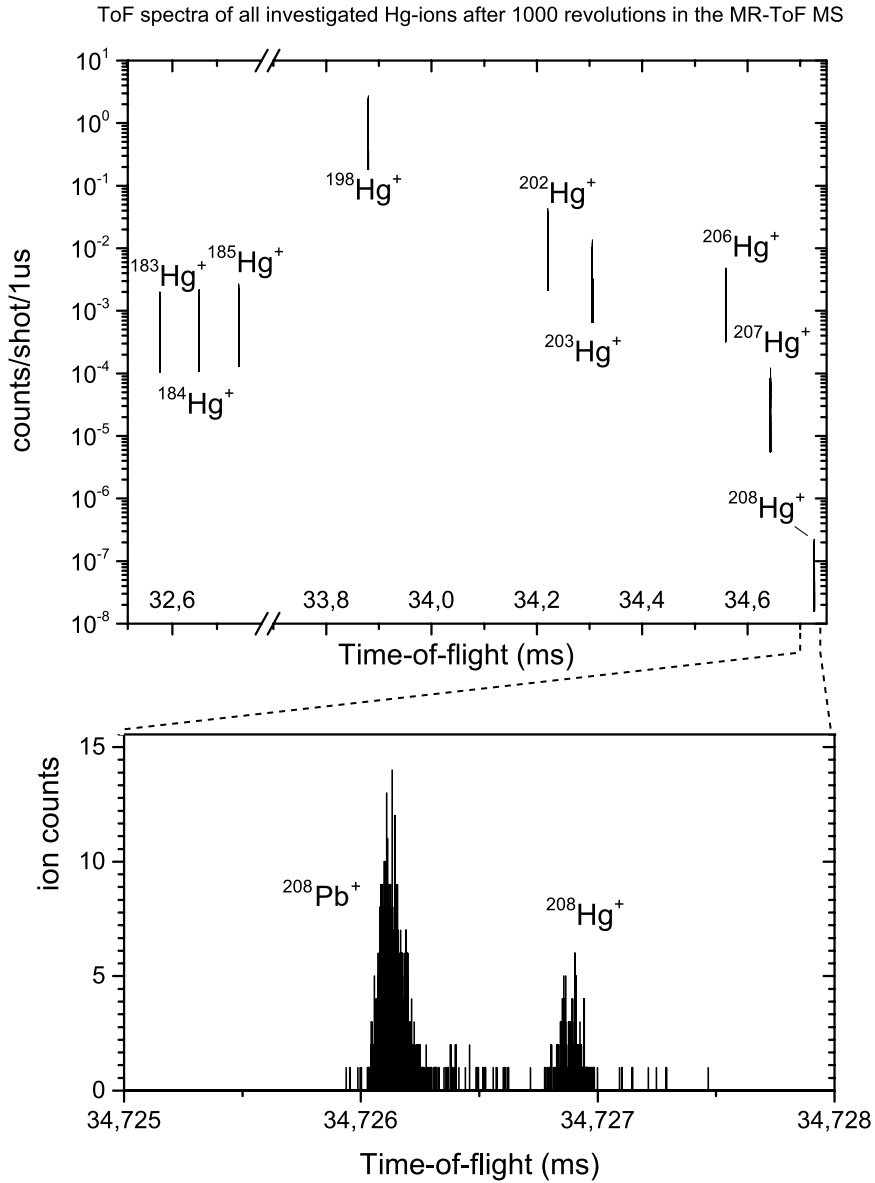


Figure 4.9: Top: Comparison of TOF versus intensity for all mercury isotopes measured with the MR-TOF setup. Bottom: Zoom in of the TOF spectrum of ^{208}Hg . Figures provided by F. Wienholtz.

4.2 Analysis

During the experiment, a total of 15 different isotopes were scanned ranging from $^{177}\text{--}^{208}\text{Hg}$. The scanned isotopes and the detection setups used for the respective isotopes are shown in Figure 4.10.

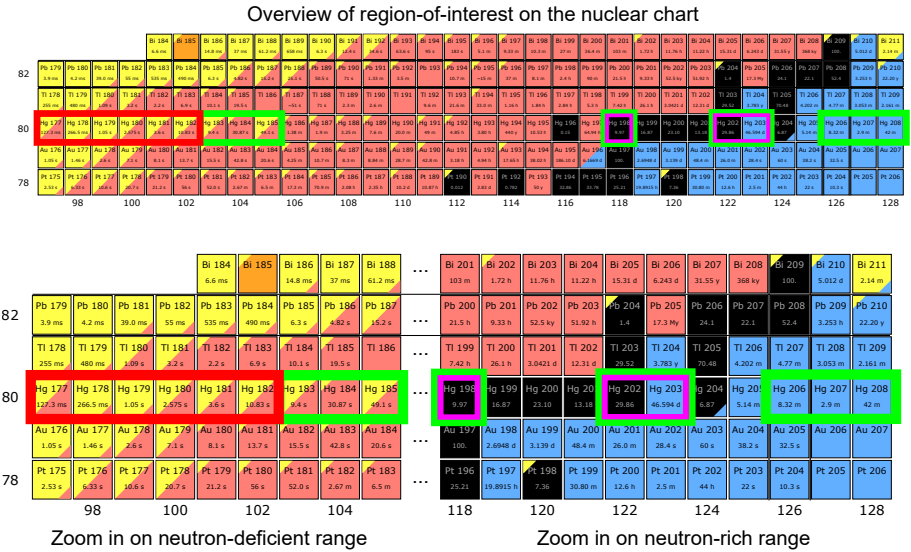


Figure 4.10: Summary of the mercury isotopes measured in the experiment, indicated by bold frames. Colored squares indicate the detection setup used for scanning. The Windmill, MR-TOF and Faraday Cup setups are indicated by red, green and pink squares respectively. Top: Overview of the total region-of-interest. Bottom: Zoom-in on neutron-deficient (left) and neutron-rich (right) parts of the isotopic chain.

4.2.1 WM data

As has been explained in section 4.1.2, decay-spectroscopy data is taken with both silicon and germanium detectors at the Windmill detection station. An overview of all spectra taken with silicon detectors Si1 and Si2, positioned at the implantation site is given in Figure 4.11. The spectra are zoomed in on the α -particle energy range and are the result of all HFS scans summed up.

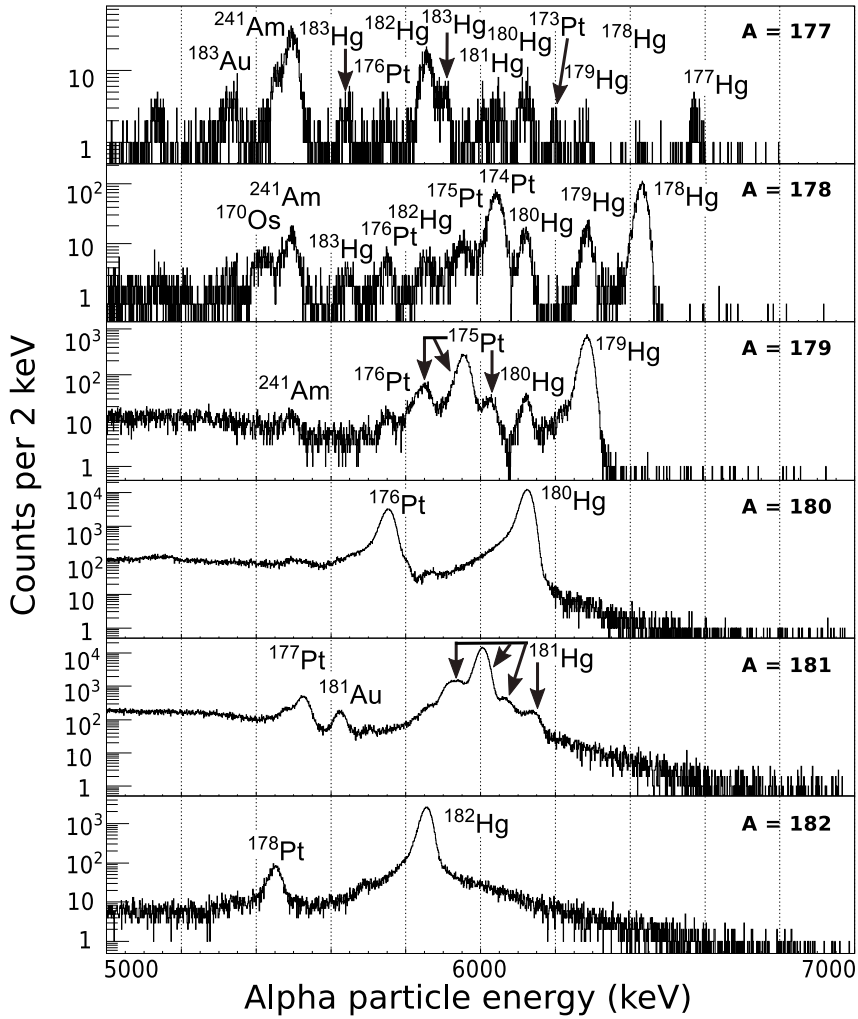


Figure 4.11: Overview of the alpha spectra obtained with Si detectors at the implantation site, summed for all HFS scans.

Similarly, γ -decay energies are recorded with the LEGe and HPGe detectors. An example of a partial spectrum taken with the LEGe-detector during a scan of ^{181}Hg is shown in Figure 4.12.

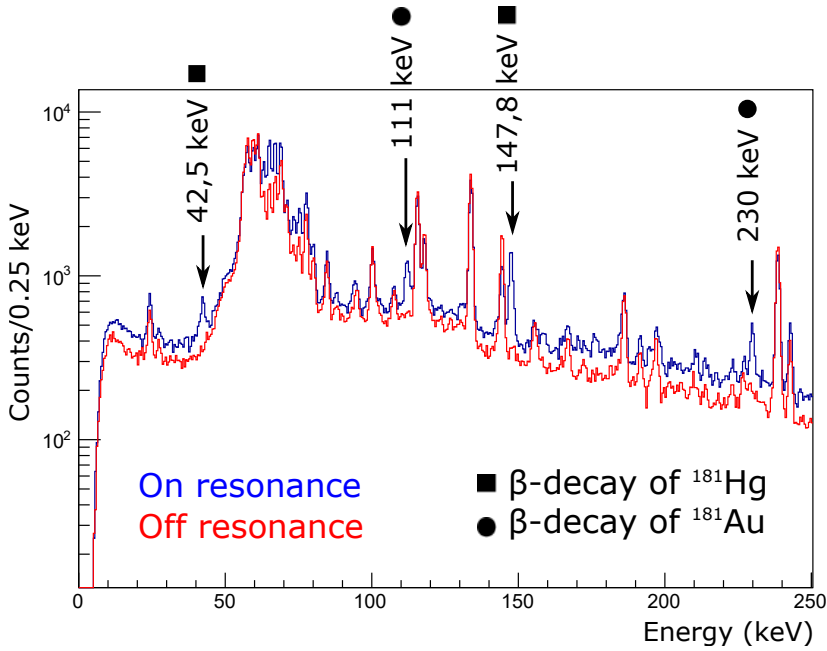


Figure 4.12: LEGe spectrum on/off resonance for a HFS scan of ^{181}Hg . The origin of gamma's with a large on/off resonance count difference are indicated.

Energy conditions for construction of the HFS

Using the mono-energetic fingerprints of α and γ -decay, it is possible to remove background contributions of contaminants by gating on detected energies related to a known decay path of the isotope under investigation. Based on the identified α peaks in Figure 4.11, energy windows were applied. The hyperfine spectra are then constructed by combining the counts recorded within these gates with the laser frequency recorded in each step. The alpha-peaks used for the HFS construction and applied energy windows are given in Table 4.2.

When constructing the HFS, only the silicon detectors from the implantation station were used as, from time to time, a SC signal was missed. This can happen when one of the veto signals, as shown in Figure 4.6, was not ready. This is for instance seen in Figure 4.14. In this figure, the irregular white gaps in the line spacing point to missed SC's such as e.g at ≈ 650 s. Moreover, adding counts of the decay-site detectors would not lead to a large increase in signal, as the

longest-lived mercury isotope, ^{182}Hg , has a half-life of only 10.8 s (see Table 4.2).

Due to the lower background and higher detection efficiency of α -particles, HFS spectra based on α -counting give rise the spectra with higher signal-to-noise ratios. When the branching ratio for α -decay is high enough, this method is preferred over γ or $\beta - \gamma$ tagging. An example is given in Figures 4.12 and 4.13, where the comparison is made for ^{181}Hg . For isotopes with lower yield, such as $^{177-179}\text{Hg}$, the count-rate was not enough to produce gamma-gated HFS spectra. Since almost all of the γ -gated spectra, except for the one shown in Fig 4.13, were of clearly lower quality than than the α -gated ones, and since they were only present for isotopes where enough statistics were already obtained via α -decay, determination of the isotope shifts and hyperfine parameters were only based on the HFS produced by α -tagging.

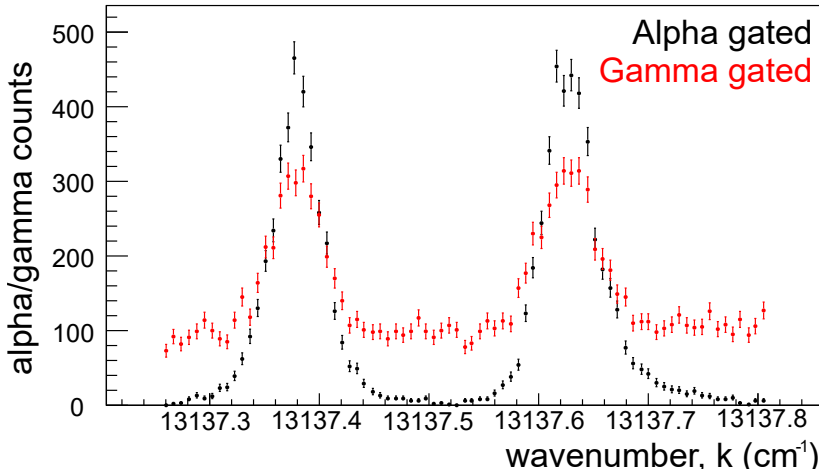


Figure 4.13: HFS scan of ^{181}Hg gating on alpha (black) and 148 keV gamma (red) energies.

Table 4.2: Summary of α -tagging contributions to the HFS measured with the WM setup and their corresponding half-lives, alpha-particle energies and intensities. Data from [95].

HFS	Isotope	$T_{1/2}$ (s)	E_α (keV)	I_α (%)	E_α -gates (keV)
^{177}Hg	^{177}Hg	0.118(8)	6579(4)	100	6500-6650
	^{173}Pt	0.382(2)	6211(6)	96(1)	6177-6217
^{178}Hg	^{178}Hg	0.266(25)	6430(6)	≥ 96	6350-6500
	^{174}Pt	0.876(19)	6039(3)	99(1)	6000-6090
^{179}Hg	^{179}Hg	1.05(3)	6285(3)	100	6200-6346
	^{175}Pt	2.54(6)	5959.5(24)	85(8)	5792-6078
			6038(10)	7.4(12)	
			5831(10)	7.3(16)	
^{180}Hg	^{180}Hg	2.58(1)	6119(4)	99.87(3)	5600-6600
			5689(5)	7.9×10^{-2}	
			5862(5)	$5.4(9) \times 10^{-2}$	
	^{176}Pt	6.3(5)	5753(3)	99.74(13)	
^{181}Hg	^{181}Hg	3.54(8)	6006(5)	87(15)	5400-6500
			5938(10)	6.1(15)	
			6071(10)	1.5(3)	
			6148(10)	0.5(1)	
	^{181}Au	14.5(3)	5626(5)	43.0(5)	
			5479(5)	43.9(5)	
	^{177}Pt	10.6(4)	5517(4)	88.5(7)	
^{182}Hg	^{182}Hg	10.83(6)	5867(5)	99	5250-6500
			5700(5)	0.57(10)	
			5455(10)	0.24(6)	
	^{182}Au	15.5(4)	5352(5)	72	
	^{178}Pt	21.1(6)	5446(3)	94.9(24)	

Deadtime correction

All obtained HFS were corrected for possible deadtime induced by very high count rates. The alpha counts recorded at each laser step are corrected for deadtime using a 100 Hz pulser. In Figure 4.14, an example of such a correction is given.

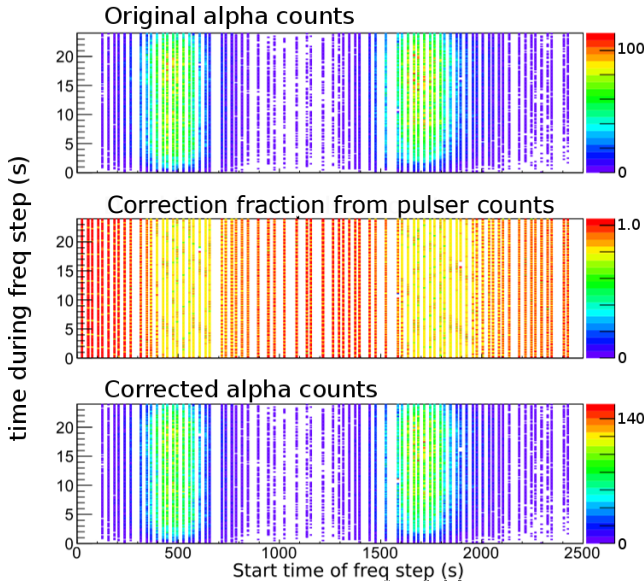


Figure 4.14: Deadtime correction factor for the example of ^{181}Hg , where the deadtime correction was the largest of all scans. Top: The original detected α counts as a function time during the laser-frequency step (y-axis) and in function of start-time of each laser step (x-axis). Middle: The fraction of recorded pulses of a 100 Hz pulser in function of time. Bottom: The same spectrum as shown on top, but now corrected for using the pulser-correction factor.

The top panel of Figure 4.14 shows the uncorrected count rate of alpha counts for ^{181}Hg for each laser step (x-axis) and as a function of time during each step (y-axis). For this particular scan, data was taken for the duration of 1 super cycle (~ 24 s) at each laser step. The middle figure shows the fraction of recorded pulser pulses, where 1 corresponds to no deadtime and 0 would correspond to no pulses recorded within a bin. The bottom figure, which represents the top figure divided by the middle one, shows the corrected counts. This method assumes that the same fraction of counts is not recorded in the silicon detector channels as for the pulser.

In Figure 4.15, the uncorrected HFS is compared with two different deadtime-correction methods. In method 1, all pulser counts recorded for a single laser step are summed up and the corresponding counts in that HFS-step are scaled according to the missing fraction of pulses. Method 2 is the result of the micro-corrections shown in Figure 4.14, where each single time bin is corrected separately. As can be seen, both methods result in almost identical HFS. All final spectra are deadtime corrected. Figure 4.15 shows the HFS, where the correction was the highest of all scans.

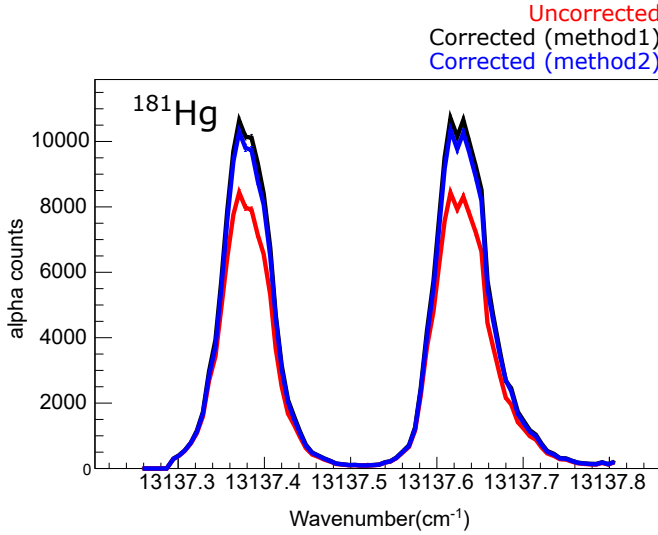


Figure 4.15: Comparison of the uncorrected (red) hyperfine spectrum of ^{181}Hg with the deadtime-corrected spectra obtained with methods 1 (black) and 2 (blue).

4.2.2 Reference measurements of ^{198}Hg

In order to check the stability in measuring the isotope and isomer shifts of a chain of isotopes, reference scans were taken using ^{198}Hg . Such scans are taken regularly between measurements in order to determine short- and long term drifts of e.g laser frequency measurements. In Figure 4.16, the time at which reference scans of ^{198}Hg were taken is shown together with the scans of different isotopes. In all but one reference measurement (measurement 7 in Fig 4.16), the scans were taken in pairs of opposite scan directions to remove potential bias.

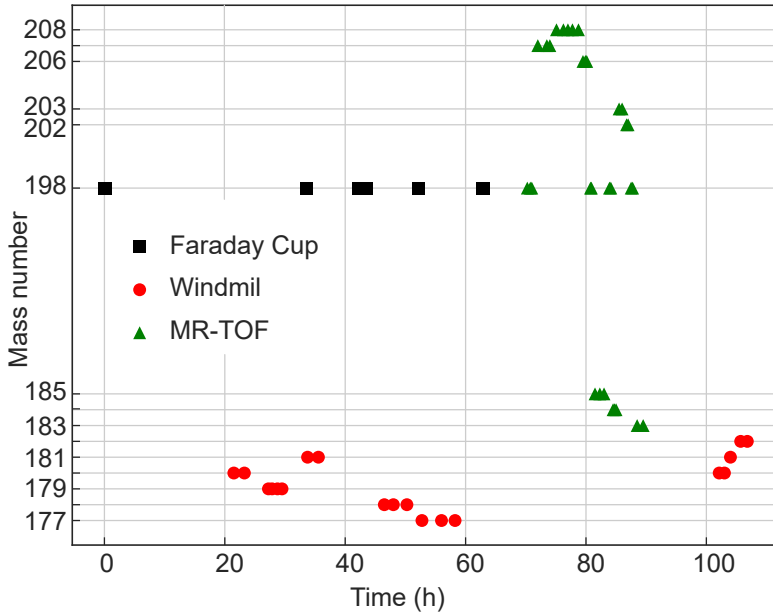


Figure 4.16: Summary of the timing of all scans performed for different mercury isotopes relative to the start of the experiment. The symbols indicate the method used for obtaining the scan.

As shown in Figure 4.16, the first 9 reference scans of ^{198}Hg were taken with the Faraday Cup at the beginning of the experiment. Due to the non-ideal separation of the GPS separator, beam contaminants of ^{197}Hg ground ($I = 1/2$) and isomeric ($I = 13/2$) states were identified in the beam. As can be seen from Figure 4.17, the corresponding HFS peaks are overlapping or even non-resolved and the full range of ^{197}Hg resonances were not covered (see e.g. left most peaks in Fig 4.17). This causes a slightly enlarged uncertainty of the peak position for the reference scans taken with the FC. When fitting the latter spectra, the hyperfine parameters a_u, b_u of the upper level were fixed to literature values [37]. At a later stage during the experiment, the reference scans were taken with the MR-TOF thereby removing the unwanted contamination.

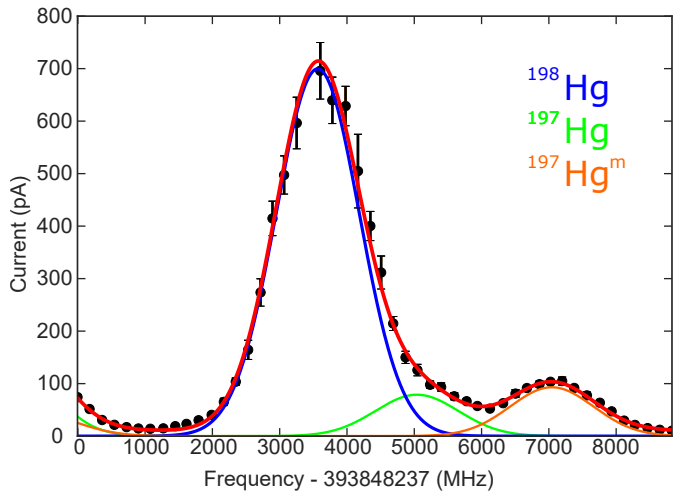


Figure 4.17: Example spectrum of ^{198}Hg Faraday Cup reference scan.

A comparison of the centroids obtained from fitting all ^{198}Hg reference scans is shown in Figure 4.18. Here, all isotope-shift reference measurements are shown with respect to the weighted average of the FC measurements. All centroids are found within a frequency range of ~ 600 MHz (~ 200 MHz fundamental frequency).

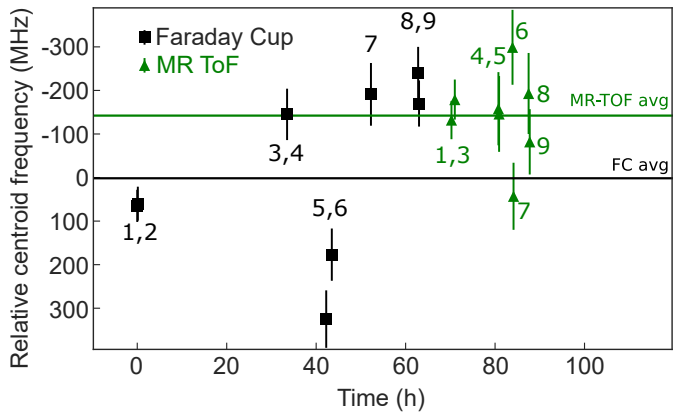


Figure 4.18: Tripled centroid frequencies of ^{198}Hg reference measurements with respect to the weighted average of the FC measurements for all FC and MR-TOF scans. MR-TOF scan number 2 is omitted due to being incomplete.

4.2.3 Fitting

In order to obtain the isotope shifts and hyperfine parameters, all scans were fitted with the Python library SATLAS [96]. This package is specifically developed for statistical data analysis of laser-spectroscopy experiments and allows for the use of both χ^2 or maximum likelihood fitting procedures. It makes use of other software packages, such as lmfit [97] for fitting and EMCEE Markov-chain Monte-Carlo method to determine the probability distributions of fit parameters from which results and errors can be extracted.

The results obtained with this package were also double-checked with results obtained with ROOT [98]. An example fit for a single scan of ^{179}Hg and the error determination using SATLAS are presented in Figure 4.19. Fits for all different isotopes measured in this experiment are shown in Figure 4.20. The final results are summarized in Tables 4.9 and 4.10.

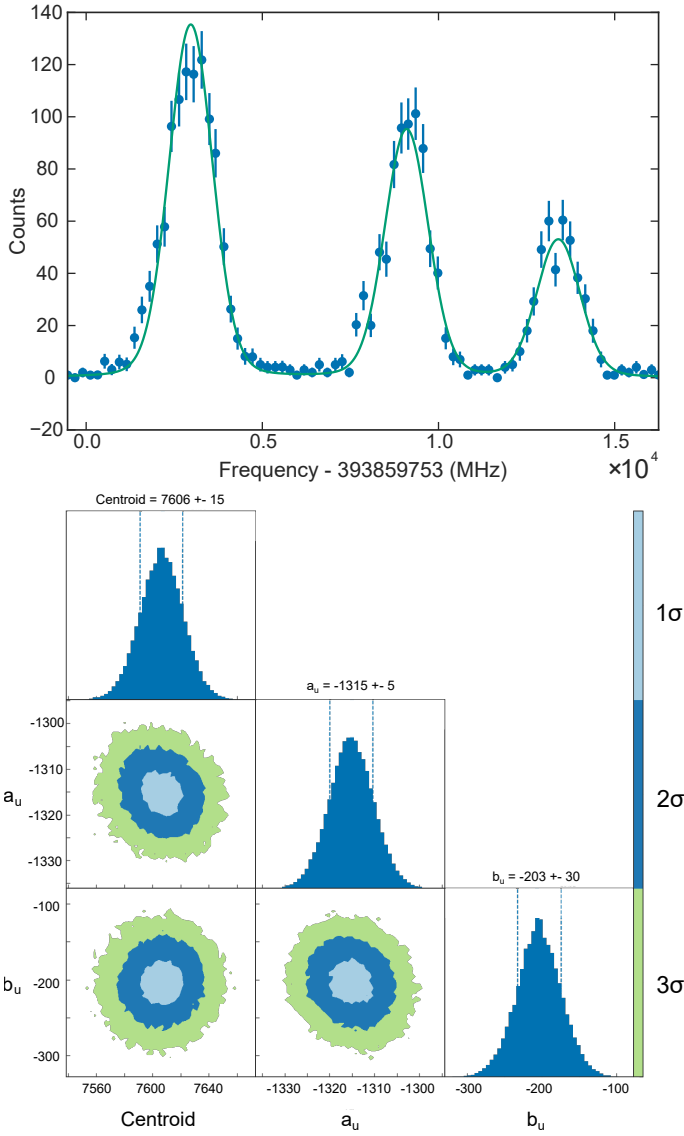


Figure 4.19: Top: Example fit for a HFS scan of ^{179}Hg . Bottom: The error analysis performed with SATLAS using the EMCEE Markov-chain Monte-Carlo method. The probability distributions for the (shifted) centroid, hyperfine a and b parameters are presented on the diagonal histograms. The 2-dimensional off-diagonal plots show the correlation between different parameters and the 1σ , 2σ and 3σ contours. Values are quoted in fundamental (non-tripled) frequencies.

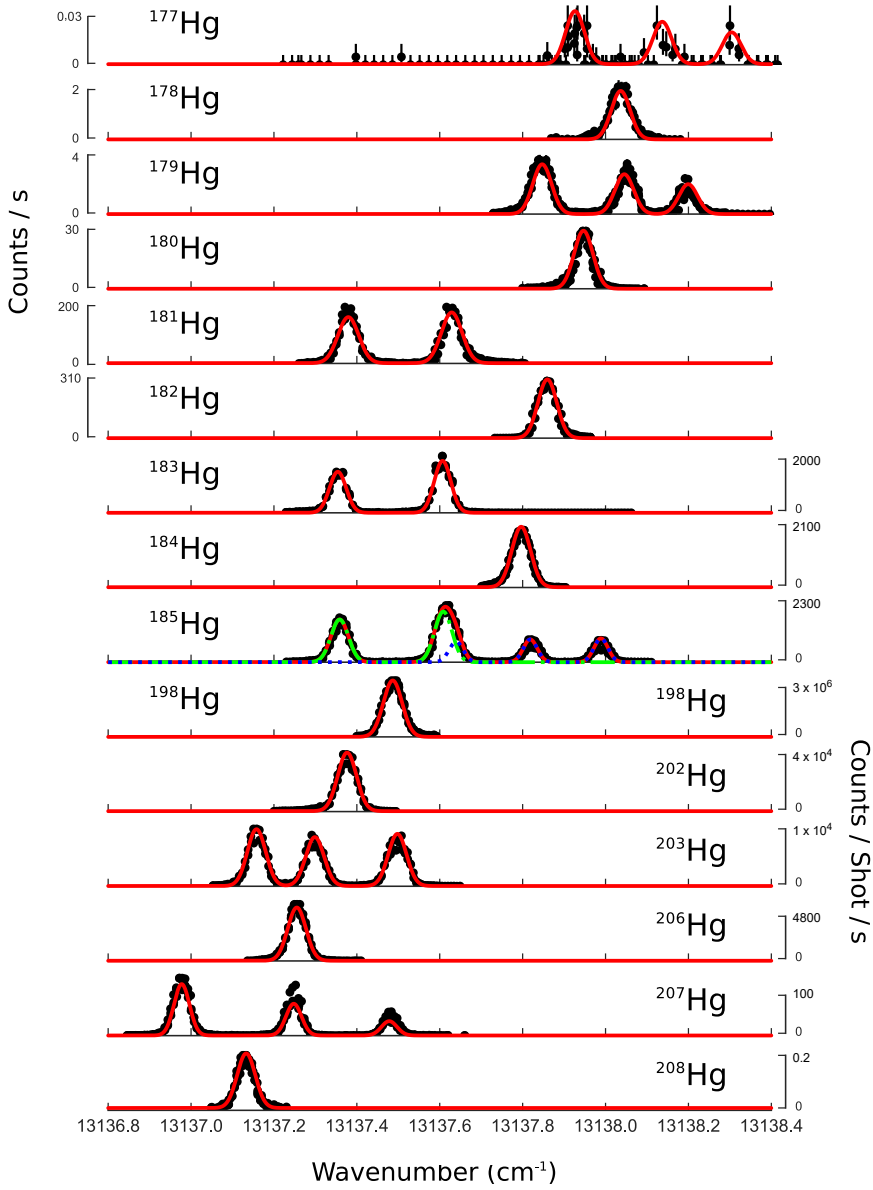


Figure 4.20: Summary of fits for all isotopes measured in this experiment. The red lines show the fit results of the data indicated by the black markers. In case of ^{185}Hg , the additional green (blue) line shows the partial fit result of the ground (isomeric) state.

4.3 Summary of results and comparison to literature

When determining the IS of different isotopes, a weighted average of several reference scans taken before and after said isotopes are taken into account. The latter was possible for all but the final measurements of $^{180,181,182}\text{Hg}$, which were taken after the last reference scan. Table 4.3 shows the reference scans that were taken into account for the IS determination in this study.

Table 4.3: Reference scans used in this study for the IS determination.

A	ref scans
177	FC 7-9
178	FC 5-9
179	FC 1-4
180 (first)	FC 1-4
180 (last)	MR-TOF avg
181 (first)	FC 3-6
181 (last)	MR-TOF avg
≥ 182	MR-TOF avg

The data of this experiment were analyzed in parallel and completely indepent by our collaborator, Dr. A. Barzakh (PNPI, Gatchina, Russia), and were compared afterwards. In his analysis, Barzakh used the mean of the two nearest reference measurements relative to the considered spectrum to determine the IS. The error on this value was chosen to cover both reference points and their individual error. A comparison of results from both analyses is shown in Tables 4.4,4.5,4.6. These tables show the good agreement between both analyses. As can be seen from Table 4.4, the IS determination is remarkably similar, considering that slightly different methods were used to take reference scans into account.

All the final values, shown in the last columns of these tables, are extracted as the average between the result of both analyses. An exception was made for ^{177}Hg , where the low statistics made a good conversion of the fitting and data extraction challenging. The fitting of Barzakh did not show such an issue, and was therefore chosen as final values.

In the analysis of this work, an increase in the IS error of ^{208}Hg is observed. This is rather unexpected, seen that the quality and count rate of the spectra were not different from the other isotopes. No such increase is seen in the analysis of Barzakh. Therefore, and in order to be consistent, the error on the

final values are based on errors obtained by Barzakh. In order to take into account the differences of the values from both analyses, half of the difference was added linearly to the final error.

Table 4.4: Comparison between IS results obtained in this analysis and that of A. Barzakh.

Isotope	IS(Barzakh) (MHz)	IS(This work) (MHz)	difference (MHz)	IS(final) (MHz)
^{177}Hg	54581(390)	54176(250)	405	54580(390)
^{178}Hg	49550(246)	49459(320)	91	49500(290)
^{179}Hg	46321(225)	46299(120)	22	46310(240)
^{180}Hg	41327(240)	41325(220)	2	41330(240)
^{181}Hg	5450(225)	5338(260)	112	5390(280)
^{182}Hg	33320(228)	33376(220)	-56	33350(260)
^{183}Hg	3119(242)	3089(280)	30	3100(260)
^{184}Hg	27635(223)	27733(275)	-98	27680(270)
^{202}Hg	-10106(175)	-10098(195)	-8	-10100(180)
^{203}Hg	-11847(180)	-11895(240)	48	-11870(200)
^{206}Hg	-20928(160)	-20933(300)	5	-20930(160)
^{207}Hg	-25813(162)	-25765(250)	-48	-25790(190)
^{208}Hg	-32040(150)	-32014(400)	-26	-32030(160)

Table 4.5: Comparison between HFS a -factor results obtained in this analysis and that of A. Barzakh.

Isotope	a (Barzakh) (MHz)	a (This work) (MHz)	difference (MHz)	a (final) (MHz)
^{177}Hg	-4320(180)	-4179(85)	-141	-4320(180)
^{179}Hg	-3986(80)	-3987(50)	1	-3990(80)
^{181}Hg	15018(105)	15048(61)	-31	15030(120)
^{183}Hg	15231(120)	15144(163)	87	15190(160)
^{203}Hg	5043(60)	5093(30)	-51	5070(90)
^{207}Hg	-4498(54)	-4500(40)	3	-4500(50)

Table 4.6: Comparison between HFS b -factor results obtained in this Analysis and that of A. Barzakh.

Isotope	$b(\text{Barzakh})$ (MHz)	$b(\text{This work})$ (MHz)	difference (MHz)	$b(\text{final})$ (MHz)
^{177}Hg	-410(600)	-414(500)	15	-410(600)
^{179}Hg	-542(200)	-562(100)	15	-550(200)
^{203}Hg	-20(200)	-12(220)	-8	-20(250)
^{207}Hg	478(210)	588(170)	-111	530(270)

4.3.1 Determination of charge radius difference and deformation

The determination of the difference in charge radius between ^{198}Hg and the isotopes measured in this work was done using the formulas detailed in 3.2 rearranged as:

$$\delta\langle r^2 \rangle^{A-198} = \left(\frac{\delta\nu^{A-198} - \left((K_{\text{NMS}} + K_{\text{SMS}}) * \left(\frac{m^A - m^{198}}{m^A * m^{198}} \right) \right)}{F * k} \right) \quad (4.1)$$

where $F = -55.36 \text{ GHz fm}^{-2}$, $K_{\text{SMS}} = (0 \pm 0.5)K_{\text{NMS}}$ with $K_{\text{NMS}} = \frac{\nu_0}{m_e/m_p}$ [5] and $k = 0.931(30)$ [21]. The results of this calculation are shown in Figure 4.21 and summarized in Table 4.10. From the data shown here, it is clear that the shape-staggering in mercury remains a localized phenomenon around the neutron mid-shell region between $N = 82$ and 126 , indicated by the dashed line with $N = 104$. A more thorough description and comparison to theoretical descriptions are given in Articles I and II. Figure 4.21 also shows that the kink in the charge radii around $N = 126$ is present in the mercury isotopic chain as well.

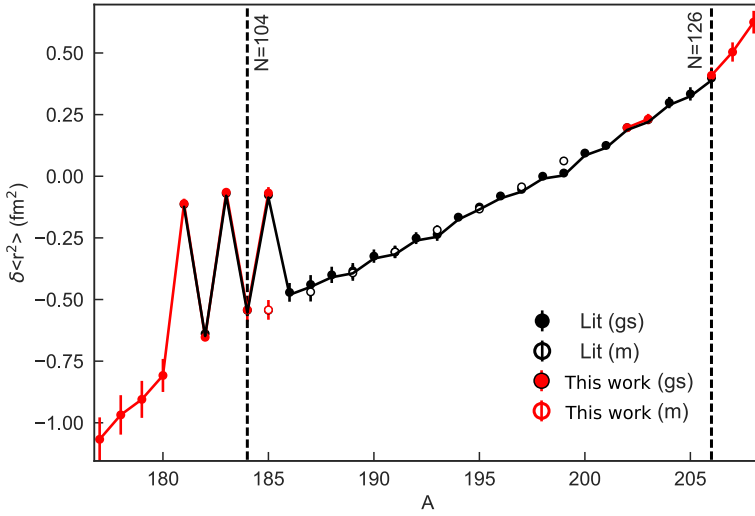


Figure 4.21: Overview of mercury charge radii differences obtained from this work and from [5]

The results of the neutron-deficient mercury isotopes and their comparison to theory is the main subject of the two papers in Section 4.4, here we will only briefly discuss the neutron-rich isotopes. For completeness, the resulting change in charge-radius gradient is given, but a more thorough analysis has been performed and will be published by T. Day Goodacre.

From Figure 4.22, focussed on the $N = 126$ region, it is clear that the characteristic kink at $N = 126$ is present in the mercury isotopic chain. Using the results from Table 4.10, the gradient in $\delta\langle r^2 \rangle$ can be obtained. The charge-radius gradient is both quantified as the ratio between the charge radius difference of the even- N isotopes on both sides of the $N = 126$ shell closure as well as for the odd- N isotopes. $R(m_{2N})$ and $R(m_{1N})$ are defined as $R(m_{2N}) = \frac{\delta\langle r^2 \rangle^{N=128,126}}{\delta\langle r^2 \rangle^{N=126,124}}$ and $R(m_{1N}) = \frac{\delta\langle r^2 \rangle^{N=127,126}}{\delta\langle r^2 \rangle^{N=126,125}}$. The results are $R(m_{2N}) = 1.980(52)$ and $R(m_{1N}) = 1.267(60)$. This result is the first experimental evidence that the kink at $N = 126$ is also present in nuclei below $Z = 82$.

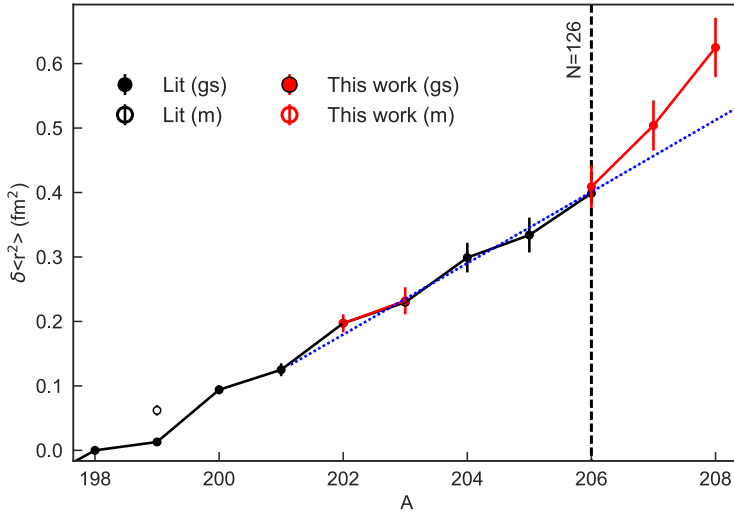


Figure 4.22: Overview of mercury charge radii differences around the $N = 126$ shell closure obtained from this work and from [5]. The blue line was added to guide the readers eye.

The magnitude of the quadrupole-deformation parameter $\langle\beta^2\rangle^{1/2}$ can be extracted from the charge-radii differences using the droplet model and a normalization via $\langle\beta^2\rangle$. The latter is extracted from a measurement of $\langle\beta^2\rangle^{1/2}$ [99], which is given as

$$\beta_2 = \frac{4\pi}{3ZR_0^2} \sqrt{B(E2)} \uparrow \quad (4.2)$$

where $B(E2)$ refers to the transition probability from the ground to the excited 2^+ states. For ^{198}Hg , $B(E2)$ was measured to be $10600(130) \text{ e}^2\text{fm}^4$, corresponding to a $\langle\beta^2\rangle^{1/2} = 0.1102(7)$. The $\langle\beta^2\rangle^{1/2}$ -deformation parameters extracted using this normalization are given in Table 4.3.1. The charge-radii are given further in Table 4.10,

From the $\langle\beta^2\rangle^{1/2}$ values shown in this table, it is seen that the mercury isotopes with $A \leq 180$, while still having a reasonable deformation, return towards a more spherical shape than the strongly-deformed $^{181,183,185}\text{Hg}$ isotopes.

Isotope	$\langle\beta^2\rangle^{1/2}$	Isotope	$\langle\beta^2\rangle^{1/2}$
^{177}Hg	0.174(20)	$^{185}\text{Hg}^m$	0.179(10)
^{178}Hg	0.182(17)	^{202}Hg	0.098(6)
^{179}Hg	0.182(17)	^{203}Hg	0.087(9)
^{180}Hg	0.190(14)	^{206}Hg	0.092(14)
^{181}Hg	0.304(2)	^{207}Hg	0.108(14)
^{182}Hg	0.197(11)	^{208}Hg	0.131(14)
^{183}Hg	0.292(2)		
^{184}Hg	0.194(9)		
^{185}Hg	0.271(2)		

Table 4.7: $\langle\beta^2\rangle^{1/2}$ -deformation parameters extracted from the charge-radii as given further in Table 4.10, normalized to ^{198}Hg with deformation parameter $\langle\beta^2\rangle^{1/2} = 0.1102(7)$.

4.3.2 Determination of nuclear moments

For the calculation of the dipole and quadrupole moment using the formulas of section 3.2, we used as reference isotopes $^{199}\text{Hg}^m$ and ^{201}Hg respectively. The spin, hyperfine a -value and magnetic dipole moment μ are , $I(^{199}\text{Hg}^m)=13/2$, $a(^{199}\text{Hg}^m) = -2298.3(2)$ MHz and $\mu(^{199}\text{Hg}^m) = -1.0147(8) \mu_N$ [100]. In the calculation of μ , the hyperfine anomaly was taken into account according to the Moskowitz-Lombardi (ML) rule [101]. This rule gives excellent description of the experimental HFA for the variety of neutron single particle states in a broad range of mercury nuclei. In this rule, the Bohr-Weisskopf contribution to the HFA is given by the simple relation

$$^{A_1}\Delta_{\text{BW}}^{A_2} = \pm\alpha \left(\frac{1}{\mu_1} - \frac{1}{\mu_2} \right), \text{ with sign from } I = l \pm \frac{1}{2} \quad (4.3)$$

where $\alpha = 1 \times 10^{-2} \mu_N$, I represents the nuclear spin, l is the orbital moment of the last neutron. The Breit-Rosentahl contribution was shown in [102] to be proportional to $\delta\langle r^2 \rangle$. Thus, $^{A_1}\Delta_{\text{BR}}^{A_2}$ can be estimated by scaling the calculated $^{201}\Delta_{\text{BW}}^{199}$ of [103]. The BW and BR contributions to the HFA for all isotopes are shown in Table 4.8. The values shown in Table 4.10 are corrected for this effect. Values for $^{181-183}\text{Hg}$ are excluded from this table due to being strongly-deformed nuclei. The ML rule requires a definite orbital quantum number l , which is undefined for such nuclei. The overall HFA, being mostly less than 1%, has only a very small effect on the outcome.

Table 4.8: BW and BR contributions to the hyperfine anomaly

A_1	$A_1\Delta_{\text{BW}}^{199} (\%)$	$A_1\Delta_{\text{BR}}^{199} (\%)$
^{177}Hg	$-0.1(50) \times 10^{-1}$	$-0.16(2)$
^{179}Hg	$0.7(50) \times 10^{-1}$	$-0.14(1)$
^{203}Hg	$2(5) \times 10^{-1}$	$0.03(1)$
^{207}Hg	$-1.7(5)$	$0.07(1)$

As shown in [104], the maximal observed deviation of the experimental HFA from the ML-calculation is equal to 4×10^{-3} . Therefore, the error to the HFA was estimated as 5×10^{-3} . This error is included in the uncertainties presented in Table 4.10. The magnetic moments and g -factors are shown in the top and bottom panel of Figure 4.23 respectively.

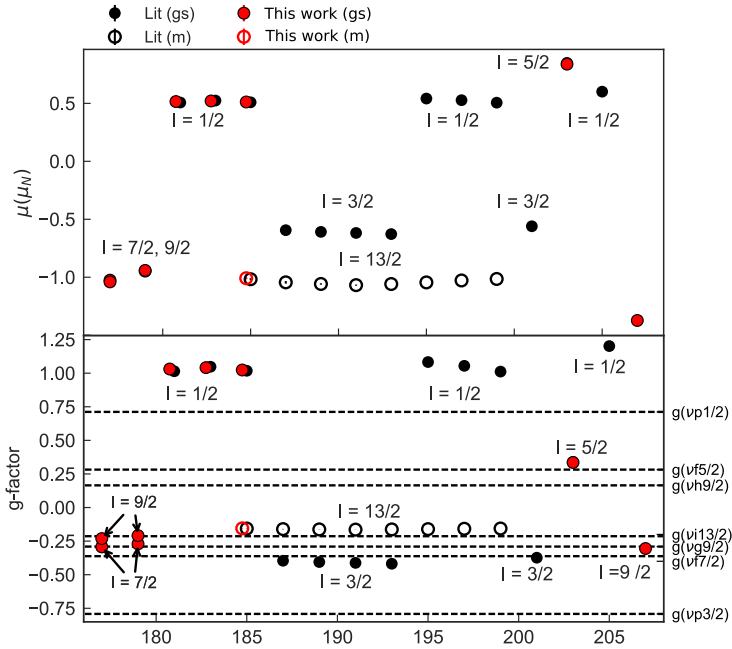


Figure 4.23: Overview of mercury magnetic moments and g -factors from this work and from [5]. The theoretical g -factor Schmidt values ($g_{\text{eff}} = 0.6g$) for odd-neutrons are shown by dashed lines.

As for $^{177,179}\text{Hg}$, only a tentative assignment of spin $I = 7/2$ had been made [105, 106], options for both $I = 7/2$ or $9/2$ are shown. From the discussion in Article II, comparing the moments of these two isotopes to other experimental moments of nuclei with a mainly $f_{7/2}$ configuration, and from the comparison with the single-particle Schmidt estimates shown for odd-neutron g -factors [57], it shows that our results agree with the previous assignment of $I = 7/2$.

^{207}Hg , on the other hand, was assumed to have a spin and parity of $I = 9/2^+$, as the neutron shell $g_{9/2}$ is the first shell above the neutron shell gap at $N = 126$. The g -factor shown at the bottom of Figure 4.23, which has been corrected with a usual $g_{\text{eff}} = 0.6$ effective g -factor, is in agreement with both the Schmidt estimate as with g -factors of other isotones with this configuration in the lead region (e.g: $g(^{209}\text{Pb}, 9/2^+) = -0.3274(4)$ n.m. [107], $g(^{211}\text{Po}, 9/2^+) = -0.266(19)$ n.m. [29]).

The used reference hyperfine b -parameter and electric quadrupole moment Q_s are $Q_s(^{201}\text{Hg}) = 0.387(6)$ b and $b(^{201}\text{Hg})$ [108, 109]. The results of both electromagnetic moment calculations are shown in Table 4.10.

Table 4.9: Summary of isotope shifts and hyperfine parameters for $6s6p^3P_1$ level in mercury and their comparison to literature.

Isotope	Spin I^π	$\delta\nu_{254nm}^{A-198}$ (MHz)	$\delta\nu_{254nm}^{A-198}$ (lit) (MHz)	a (MHz)	a (lit) (MHz)	b (MHz)	b (lit) (MHz)
^{177}Hg	$(7/2^-)^\dagger$ $(9/2^-)^\dagger$	54580(390) 55170(390)	- -	-4320(180) -3460(180)	- -	-414(600) -875(600)	- -
^{178}Hg	0^+	49500(290)	-	-	-	-	-
^{179}Hg	$(7/2^-)^\dagger$ $(9/2^-)^\dagger$	46310(240) 46820(230)	- -	-3990(80) -3147(72)	- -	-554(200) -1050(210)	- -
^{180}Hg	0^+	41330(240)	-	-	-	-	-
^{181}Hg	$1/2^-$	5390(280)	5560(200)	15030(120)	14960(250)	-	-
^{182}Hg	0^+	33350(260)	-	-	-	-	-
^{183}Hg	$1/2^-$	3110(260)	3310(100)	15190(160)	15380(30)	-	-
^{184}Hg	0^+	27690(270)	27720(90)	-	-	-	-
^{185}Hg	$1/2^-$	3300(600)	3710(30)	14928(340)	14960(70)	-	-
^{185m}Hg	$13/2^+$	27780(500)	27770(110)	-2288(11)	-2305(19)	110(300)	-140(230)
^{202}Hg	0^+	-10100(180)	-10102.4(4.2)	-	-	-	-
^{203}Hg	$5/2^-$	-11870(200)	-11750(180)	5043(86)	4991.33(4)	-20(250)	-249.2(3)
^{206}Hg	0^+	-20930(160)	-20420(80)	-	-	-	-
^{207}Hg	$(9/2^+)^\dagger$	-25790(190)	-	-4499(56)	-	533(260)	-
^{208}Hg	0^+	-32030(160)	-	-	-	-	-

 † Assumed nuclear spin

Table 4.10: Summary of resulting mean-square charge differences ($\delta\langle r^2 \rangle^{A-198}$) and nuclear moments (μ , Q) and their comparison to literature.

Isotope	Spin I^π	$\delta\langle r^2 \rangle^{A-198}$ (fm ²)	$\delta\langle r^2 \rangle^{A-198}$ (lit) (fm ²)	μ (μ_N)	μ (lit) (μ_N)	Q_s (b)	Q_s (lit) (b)
^{177}Hg	$(7/2^-)$	-1.067(8){78}	-	-1.025(40)	-	0.57(83)	-
	$(9/2^-)$	-1.083(8){78}	-	-1.040(50)	-	1.21(91)	-
^{178}Hg	0^+	-0.968(6){71}	-	-	-	-	-
^{179}Hg	$(7/2^-)$	-0.905(5){70}	-	-0.948(20)	-	0.76(28)	-
	$(9/2^-)$	-0.915(5){70}	-	-0.941(27)	-	1.45(31)	-
^{180}Hg	0^+	-0.808(5){60}	-	-	-	-	-
^{181}Hg	$1/2^-$	-0.111(6){11}	-0.114(4)	0.515(4)	0.5071(7)	-	-
^{182}Hg	0^+	-0.653(5){10}	-0.6384(20)	-	-	-	-
^{183}Hg	$1/2^-$	-0.065(5){7}	-0.0694(19)	0.521(6)	0.524(5)	-	-
^{184}Hg	0^+	-0.542(5){40}	-0.5436(22)	-	-	-	-
^{185}Hg	$1/2^-$	-0.069(6){7}	-0.0764(6)	0.512(2)	0.509(4)	-	-
^{185m}Hg	$13/2^+$	-0.543(4){40}	-0.5433(22)	-1.008(10)	-1.017(9)	-0.15(41)	0.19(32)
^{202}Hg	0^+	0.197(3){14}	0.19728(23)	-	-	-	-
^{203}Hg	$5/2^-$	0.232(5){17}	0.2296(35)	0.843(15)	0.84895(13)	0.03(35)	0.40(4)
^{206}Hg	0^+	0.409(3){30}	0.3987(16)	-	-	-	-
^{207}Hg	$(9/2^+)$	0.503(4){37}	-	1.373(20)	-	-0.73(37)	-
^{208}Hg	0^+	0.625(3){45}	-	-	-	-	-

Literature values shown of recalculated IS from A and B values in [5].

For determination of β_2 , the parametrization of Berdichevsky and Tondetur was applied [52].

Statistical errors are given in parenthesis. Systematic errors stemming from

the indeterminacy of the F factor (7%), see [5] and M_{SMS} are shown in curly brackets

Ref. for μ : $\mu(^{199}\text{Hg}^m) = -1.0147(8) \mu_N$, $A(^{199}\text{Hg}^m) = -2298.3(2) \text{ MHz}$ [100]

Ref. for Q_s : $Q_s(^{201}\text{Hg}) = 0.387(6) b$ [109], $B(^{201}\text{Hg}) = -280.107(5) \text{ MHz}$ [108]

4.4 Publications (Articles I and II)

The following articles deal with the results of the mercury isotopes obtained in the in-source laser spectroscopy experiment detailed in this chapter. In both articles, only the neutron-deficient $^{177-186}\text{Hg}$ isotopes are discussed.

By measuring the IS and HFS parameters, and deducing the charge-radii differences and electromagnetic moments, it is firmly established that the large odd-even staggering is a localized phenomenon situated around the neutron mid-shell between $A = 180$ and $A = 186$. Both the charge radii and electromagnetic moments suggest small deformations for mercury isotopes with $A \leq 180$. The magnetic moments of $^{177,179}\text{Hg}$ seem to agree well with the previous tentative assignments of $7/2^-$.

The results were compared with theoretical calculations using both mean-field DFT and Monte-Carlo Shell Model methods. The DFT calculations (e.g. using a UNEDF1^{SO} and SLy5sX) qualitatively predict a large jump in charge radius in the vicinity of the neutron mid shell $N = 104$. Both calculations use a very different tuning parameter (pairing and surface tension respectively) in order to come to a similar result. The emergence of several close-lying minima in the energy surface inhibits the determination of the correct staggering amplitude and location along the chain of isotopes.

Results from MCSM calculations show an excellent agreement with experiment and additionally provide a microscopic understanding of the origin of the shape staggering. These calculations are compared to both the charge-radii differences and to the electromagnetic moments. This was the first time that such calculations were performed in such a heavy system where 30 protons and up to 24 neutrons were interacting in a very large model space. These calculations explain shape-staggering in mercury as resulting from a rearrangement of nucleon occupation numbers due to the monopole interaction between the $\pi(h_{9/2})$ and $\nu(i_{13/2})$ nucleon orbits, and by doing so, provide a microscopic insight in this phenomenon. This responsible mechanism links the origin of the shape staggering with other interesting nuclear structure phenomena across the nuclear chart, such as for instance the quantum phase transition in the zirconium isotopes or shape coexistence in the nickel nuclei [110,111].

Mercurial nuclei get back in shape

B. A. Marsh^{1,*}, T. Day Goodacre^{1,2,+}, S. Sels^{3,+}, Y. Tsunoda⁴, B. Andel⁵, A. N. Andreyev⁶, N. A. Althubiti², D. Atanasov⁷, A. E. Barzakh⁸, J. Billowes², K. Blaum⁷, T. E. Cocolios², J. G. Cubiss⁶, J. Dobaczewski⁶, G. J. Farooq-Smith², D. V. Fedorov⁸, V. N. Fedosseev¹, K. T. Flanagan², L. P. Gaffney^{3,9}, L. Ghys³, M. Huyse³, S. Kreim⁷, D. Lunney¹⁰, K. M. Lynch¹, V. Manea⁷, Y. Martinez Palenzuela³, P. L. Molkanov⁸, T. Otsuka^{3,4,11,12,13}, A. Pastore⁶, M. Rosenbusch^{12,14}, R. E. Rossel¹, S. Rothe^{1,2}, L. Schweikhard¹⁴, M. D. Seliverstov⁸, P. Spagnoletti⁹, C. Van Beveren³, P. Van Duppen³, M. Veinhard¹, E. Verstraelen³, A. Welker¹⁵, K. Wendt¹⁶, F. Wienholtz¹⁴, R. N. Wolf⁷, A. Zadvornaya³, and K. Zuber¹⁵

¹CERN, Geneva, Switzerland

²School of Physics Astronomy, The University of Manchester, Manchester M13 9PL, UK

³KU Leuven, Instituut voor Kern- en Stralingsfysica, B-3001 Leuven, Belgium

⁴Center for Nuclear Study, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

⁵Department of Nuclear Physics and Biophysics, Comenius University in Bratislava, 84248 Bratislava, Slovakia

⁶University of York, Department of Physics, YO10 5DD, York, UK

⁷Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

⁸Petersburg Nuclear Physics Institute (PNPI), NRC Kurchatov Institute, Gatchina 188300, Russian Federation

⁹School of Engineering and Computing, University of the West of Scotland, High Street, Paisley, PA1 2BE, UK

¹⁰CSNSM-IN2P3-CNRS, Université Paris-Sud, 91406 Orsay, France

¹¹Department of Physics, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

¹²RIKEN Nishina Center, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

¹³National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, Michigan 48824, USA

¹⁴Ernst-Moritz-Arndt-Universität, Institut für Physik, 17487 Greifswald, Germany

¹⁵Technische Universität Dresden, 01069 Dresden, Germany

¹⁶Johannes Gutenberg Universität 55099 Mainz, Germany

*bruce.marsh@cern.ch

+these authors contributed equally to this work

ABSTRACT

Atomic nuclei exhibit single-particle and collective degrees of freedom, making them susceptible to variations in size and shape when adding or removing nucleons. The rare cases where dramatic changes in shape occur with the removal of only a single nucleon are key for pinpointing the components of the nuclear interaction driving nuclear deformation. Laser spectroscopy probes the nuclear charge distribution, revealing attometer-scale variations and highlighting sensitivity to the proton (Z) and neutron (N) configurations of the nucleus. The lead isotopes, which possess a closed proton shell ($Z = 82$), are spherical and steadily shrink with decreasing N . A surprisingly different story was observed for their close neighbours, the mercury isotopes ($Z = 80$) almost half a century ago^{1,2}: Whilst the even-mass isotopes follow the trend seen for lead, the odd-mass isotopes ^{181,183,185}Hg exhibit a striking increase in charge radius. This dramatic ‘shape staggering’ between even- and odd-mass isotopes remains a unique feature of the nuclear chart.

Here we present the extension of laser spectroscopy results that reach ¹⁷⁷Hg. An unprecedented combination of state-of-the-art techniques including resonance laser ionization, nuclear spectroscopy and mass spectrometry, has established ¹⁸¹Hg as the shape-staggering endpoint. Accompanying this experimental tour de force, recent computational advances incorporating the largest valence space ever used have been exploited to provide Monte-Carlo Shell Model calculations, in remarkable agreement with the experimental observations. Thus, microscopic insight into the subtle interplay of nuclear interactions that give rise to this phenomenon has been obtained, identifying the shape-driving orbitals. Although shape staggering in the mercury isotopes is a unique and localized feature in the nuclear chart, the underlying mechanism that has now been uncovered nicely describes the duality of single-particle and collective degrees of freedom in atomic nuclei.

Atomic nuclei, comprising protons and neutrons, display a rich array of quantum phenomena. These complex many-body systems obey the Pauli exclusion principle which dictates a shell-like structure, akin to Bohr’s model of the atom.

In the vicinity of closed shells, at the ‘magic numbers’ of $Z, N = 8, 20, 28, 50, 82$ and $N = 126$, the nuclear wavefunction is dominated by the last few particles (or holes) and excitations thereof. In contrast to this single-particle nature,

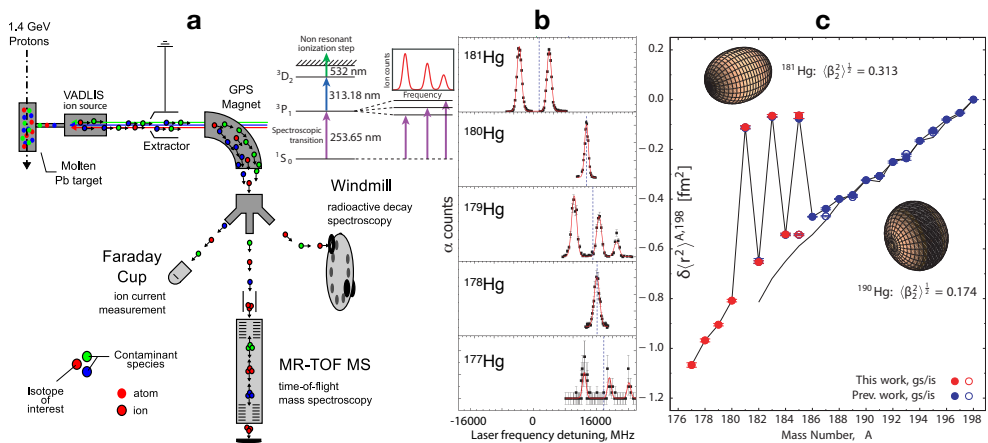


Figure 1. Schematic of the in-source resonance ionization spectroscopy method for the study of radiogenic mercury isotopes (see Methods) (a): mercury isotopes are produced by proton-induced nuclear reactions in a molten lead target. The vapor effuses into the anode volume of the VADLIS ion source³ where the atoms are ionized using a 3-step resonance photo-ionization scheme (inset). The ions are extracted as a mono-energetic beam at 30 keV. The isotope of interest is selected using the general-purpose mass separator (GPS) and directed towards the most appropriate of the three detection systems depicted. By scanning the 1st-step laser wavelength, the isotope shift (IS) and hyperfine structure (HFS) is studied (b). The isotope shifts are used to calculate the changes in mean-squared charge radii $\delta\langle r^2 \rangle$ with respect to $N = 126$ along the isotopic chain (c). The results of this study appear as red circles. ^{177–180}Hg are new measurements, while ^{181–185}Hg were re-measured and the data points overlap with those of the literature values, as can also be seen by the close agreement between values in Table 1. The error bars indicate the statistical uncertainty corresponding to the measurement precision. The scaling uncertainties arising from the indeterminacy of the F factor (7%⁴) and the Specific Mass Shift, M_{SMS} are discussed in the Methods section and provided in Table 1 (see Supplementary Information linked to the on-line version). The additional black line illustrates the previously measured quasi-spherical trend of the lead nuclei⁵.

collective behaviour appears away from the closed shells, as increased nucleon-nucleon correlations drive the minimum-energy configuration of the nucleus to deformation. Consequently, the ground-states of most isotopes in the nuclear chart have non-spherical shapes, the most common being the prolate (rugby-ball) shape. Different shapes, corresponding to alternative nucleon configurations, can coexist within the same nucleus^{6,7}. It remains a challenge to present the full picture of the underlying microscopic origin of this phenomenon.

Optical spectroscopy is able to measure subtle shifts in the energy of the atomic electron levels, arising from changes in the charge distribution of the nucleus⁸. Along the isotopic chain of a given element, this effect is known as the isotope shift. From this, the change in mean-square charge radius, $\delta\langle r^2 \rangle$, can be extracted in a nuclear-model independent way. Similarly, the hyperfine splitting of the electronic levels gives direct access to the nuclear properties: spin (I), magnetic dipole (μ) and electric quadrupole (Q) moments. Such measurements are therefore a sensitive and direct probe of the valence particle configuration and changes in nuclear

size or deformation as a result of the addition, removal, and consequential redistribution of nucleons.

The radioactive isotopes in the lead region have been the subject of a variety of optical spectroscopy studies for several decades. An intensified interest in this region was sparked by the study of the mercury isotopic chain in which a sudden and unprecedented increase in charge radius was observed for ¹⁸⁵Hg, ¹⁸³Hg and ¹⁸¹Hg^{1,2}. For the heavier mercury isotopes the changes in charge radii mirror those of lead⁵: steadily shrinking with decreasing N . This seminal discovery of shape-staggering between odd and even mercury isotopes is unparalleled elsewhere in the nuclear chart and was key to establishing the idea of shape coexistence at low excitation energy^{7,9}. A plethora of studies on the excited states of these nuclei^{10,11} provided a further substantial insight into shape-coexistence, complementing the laser spectroscopy studies of ground and isomeric-states. However, to acquire a full understanding of this phenomenon two challenges remain:

Understanding the nature of this spectacular occurrence requires its precise localisation by probing even more neutron-deficient systems, which were experimentally inaccessible.

Likewise, theoretical solutions have been thwarted by the enormous computational requirements of a fully microscopic, many-body calculation of such heavy systems.

In this Letter we report breakthroughs on both fronts that now provide explanations of the underlying mechanism and localized nature of shape coexistence:

1. The combination of state-of-the-art radioisotope production and detection techniques (see Figure 1a) at the CERN-ISOLDE radioactive ion beam facility¹², extending laser spectroscopy measurements to four lighter mercury isotopes; and
2. The exploitation of recent advances in computational physics to perform Configuration-Interaction Monte Carlo Shell Model (MCSM)¹³ calculations with the largest model spaces to date.

The new experimental scheme is illustrated in Fig. 1a with the recorded laser spectra shown in Fig. 1b and resulting charge-radii data in Fig. 1c.

Ground-state charge radii and magnetic moments were studied by performing resonance ionization spectroscopy on the mercury isotopes with unprecedented sensitivity, reaching as far as ¹⁷⁷Hg, an isotope with a half-life of only 127 milliseconds and a production rate as low as one ion per minute.

The isotope shifts measured relative to ¹⁹⁸Hg and the deduced changes in mean-square charge radii and moments for ^{177–185}Hg are presented in Table 1 (see Supplementary Information linked to the on-line version). The changes in mean-square charge radii of mercury isotopes combined with those of the lead isotopes⁵ are shown relative to $N = 126$ in Figure 1c. In addition to confirming the earlier results, these new data firmly prove that the dramatic shape staggering is a local phenomenon and that the odd-mass mercury isotopes return to sphericity at $A = 179$ ($N = 99$).

In order to pinpoint the microscopic origin of this observation, we performed large-scale numerical simulations for the quantum many-body problem using a MCSM method. Heavy nuclei such as the mercury isotopes are beyond the limit of conventional Configuration Interaction (CI) calculations for protons and neutrons interacting through nuclear forces. However, using the MCSM method and the most advanced computers has allowed us to reach this region for the first time, redefining the state of the art of CI calculations for atomic nuclei. Calculations were performed for the ground and the lowest excited states in ^{177–186}Hg with spin and parity corresponding to the experimental ground-state values of the nuclei considered: 0^+ for the even-mass isotopes, $1/2^-$ for ^{181,183,185}Hg and $7/2^-$ for ^{177,179}Hg. Also the $13/2^+$ long-lived isomer in ¹⁸⁵Hg was examined. The MCSM provides the eigen wavefunction from which the magnetic dipole and electric quadrupole moments, and the shape parameters (β_2 , γ) are calculated¹⁴. In turn, the shape parameters are used to obtain the mean-square charge radius (see Methods). For all these states, the changes in mean-square charge radii relative to the ground state of ¹⁸⁶Hg are presented as shaded boxes

in Figure 2a. The height of the shaded box is related to the spread of the quantum fluctuations of the MCSM eigenstates (see Methods). The figure shows that for every mercury isotope, only one state can be identified whose calculated $\delta\langle r^2 \rangle$ is in agreement with experiment. For the odd-mass nuclei this identification is confirmed by the agreement between the calculated and measured magnetic moments, as shown in Figure 2b. Furthermore, with the exception of ¹⁸¹Hg and in spite of the limited number of basis vectors used, the selected MCSM states correspond indeed to the ground states in the calculations. The remarkable agreement with the experimental data that has been achieved reinforces our confidence in ability of these MCSM calculations to reveal the discrete changes in the nuclear configuration that drive shape coexistence in this region. We therefore gain an insight into the underlying mechanism responsible for the sudden appearance and disappearance of the uniquely-pronounced shape staggering in the mercury isotopes.

The MCSM enables us to examine the microscopic composition of each MCSM state in terms of the occupancy of the proton and neutron orbitals (see Figure 2c). The most striking differences between the deformed $1/2^-$ states in ^{181,183,185}Hg and the other near-spherical states are found in the nucleon-occupancy of two orbitals: the proton $1h_{9/2}$ situated above the $Z = 82$ closed shell and the neutron $1i_{13/2}$ midshell between $N = 82$ and $N = 126$. The strongly deformed $1/2^-$ states of ^{181,183,185}Hg exhibit large and constant values (~ 8) of the neutron $1i_{13/2}$ occupation number, as well as a sizable promotion of 2 to 3 protons across the $Z = 82$ magic shell gap to the $1h_{9/2}$ orbit.

This abrupt and significant reconfiguration of the nucleons originates not only from the quadrupole component of the nucleon-nucleon (NN) interaction, known to be responsible for inducing deformation in atomic nuclei, but also in the monopole component. The strong attractive nature of the latter, specifically between the proton $1h_{9/2}$ orbital and neutron $1i_{13/2}$ orbital, results in an additional lowering of the binding energy¹⁴. If this energy gain, combined with that of the quadrupole deformation, exceeds the energy needed to create particle-hole excitations then the deformed state becomes the ground state.

It is thus the combined action of the monopole interaction, whose effect depends linearly on the orbital occupancy numbers, and the quadrupole interaction, that follows a quadratic dependence with a maximum when an orbital is half-filled (i.e. 7 neutrons in the $1i_{13/2}$ orbital) that delineates the deformed region for the mercury isotopic chain between $N = 101$ and $N = 105$. For even- N mercury isotopes, the pairing correlation that exists for the spherical shape produces sufficient binding energy because of the high level density, whereas this is suppressed in odd- N isotopes due to blocking of the unpaired neutron. The strongly deformed state, assisted by the combined monopole and quadrupole effect, therefore becomes the ground state. Thus, the observed shape staggering is due to a subtle competition between these two

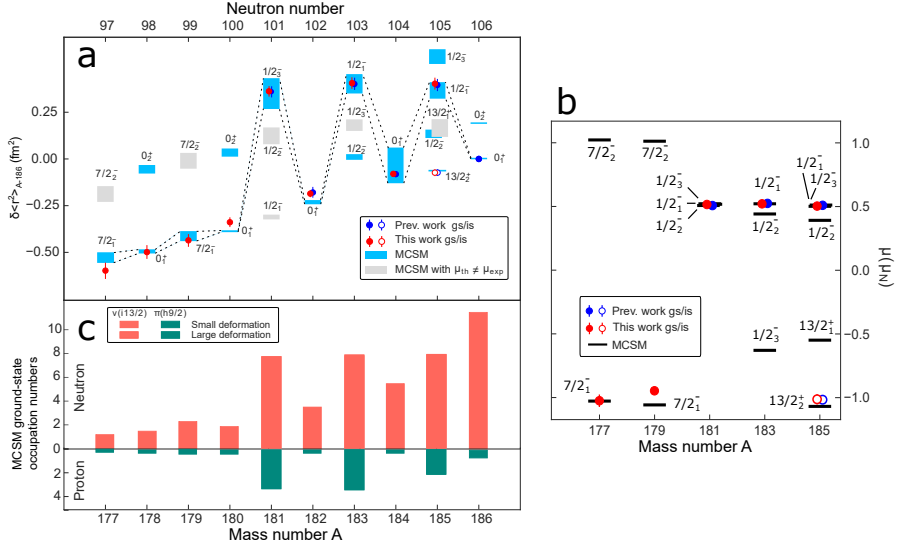


Figure 2. **a:** The upper left panel shows $\langle r^2 \rangle$ relative to that of the ground state of ^{186}Hg . Red points are experimental data from this experiment. They include the combined statistical and systematic uncertainties quoted in Table 1. Blue points refer to literature values from⁴. The shaded boxes indicate radii corresponding to the MCSM eigenstates labeled by their respective spin J , parity π and energetic ordering i as J_i^π . The gray areas show MCSM eigenstates for which the calculated magnetic moment differs from measured value. **b:** Comparison of the calculated and experimental magnetic moments for different states in the odd-mass mercury isotopes. **c:** The occupation numbers of the neutron $i_{13/2}$ orbit and the proton $h_{9/2}$ orbit for the states displayed by the blue connected areas in panel **a** (these are the experimentally-observed ground-states). The deviation of the three strongly prolate deformed states at $A = 181, 183, 185$ from the general occupancy trend is evident.

shapes, which are different in terms of their many-body configuration. The shape staggering effect manifests characteristic features of a quantum phase transition^{15–17}: In a given nucleus, different phases - a near spherical and a strongly deformed nuclear shape - appear at almost the same energy without mixing. By making small changes in the control parameter, which in this case is the neutron number, the system alternates between the two phases. In the case of the mercury chain, this observation, which resembles a critical phenomenon in phase transitions, happens six times. This unique feature is now quantitatively understood through the MCSM calculations, highlighting the importance of the simultaneous multiple excitations to the proton $1h_{9/2}$ and neutron $1i_{13/2}$ orbitals¹⁴. In this picture, because of the major role of the neutron $1i_{13/2}$ orbital in driving the large quadrupole deformation, this effect can only dominate close to neutron number $N=103$. By delineating the region of shape staggering our new data provides the necessary information to support this picture.

In summary, the lightest mercury isotopes return towards sphericity and the region of shape staggering is delineated

between $N = 101$ and $N = 105$. Its underlying microscopic origin has been identified through MCSM calculations as an abrupt and significant reconfiguration of the nucleon orbital occupancies.

The extension of the MCSM calculations to other states should allow further comparisons with available experimental information on the different band structures identified in nuclear spectroscopy studies. This will be complemented by information from Coulomb excitation and nucleon transfer experiments on the ground and isomeric states, now possible using post-accelerated radioactive ion beams¹⁸.

In this work we have uncovered the general mechanism that links different features in various locations of the nuclear chart, thereby providing a deeper understanding of the structure of atomic nuclei in general.

Methods summary

The experiment was performed at the CERN-ISOLDE isotope separator facility using the technique of in-source resonance ionization spectroscopy¹⁹, as depicted in Figure 1.

Mercury isotopes were produced by spallation of lead nuclei in a liquid target using up to $1\ \mu\text{A}$ of $1.4\ \text{GeV}$ protons from the CERN PS-Booster Synchrotron. The radiogenic mercury vapor diffuse into the anode cavity of the Versatile Arc Discharge and Laser Ion Source (VADLIS) where resonance ionization spectroscopy was performed by wavelength-tuning of the $254\ \text{nm}$ first-step transition ($6s^2\ ^1\text{S}_0 \rightarrow 6d6p\ ^3\text{P}_1$)[◦] of the 3-step ionization scheme for Hg^+ ion production (Figure 1a inset)²⁰.

The ion beam was mass separated using a magnetic-dipole spectrometer, set to transmit a chosen isotope of interest to the most suitable of three detection systems, depending on the beam intensity, purity, decay mode and half-life of the species: the ISOLDE Faraday cup ($>100\ \text{fA}$); the ISOLTRAP MR-ToF MS²¹ (isobar separated ion counting of longer lived isotopes); or the Windmill decay spectroscopy setup (for efficient and selective detection of alpha-decaying isotopes). Laser spectra were obtained for all isotopes from $^{185}\text{--}^{177}\text{Hg}$. Isotope shifts, relative to the reference isotope ^{198}Hg , and hyperfine parameters were measured. From these observables, the changes in mean-square charge radii together with spins and magnetic moments were deduced.

Large-scale Monte Carlo shell-model calculations (MCSM) were performed with neutron-neutron and proton-proton interactions taken from²² and with the proton-neutron interaction taken from²³. Single-particle energies were varied smoothly as functions of Z and N between the respective values of doubly-magic ^{132}Sn and ^{208}Pb . The Hamiltonian was fixed and kept throughout this study. Effective charges for protons and neutrons were used, with their actual values being $1.6e$ and $0.6e$ respectively, together with a spin quenching factor of 0.9 . The model space in which the nucleons interact are 11 proton orbits from $1g_{7/2}$ up to $1i_{13/2}$ and 13 neutron orbits from $1h_{9/2}$ up to $1j_{15/2}$, corresponding to the largest scale MCSM calculation ever performed.

Methods

Mercury isotope production

The experiment was performed at the CERN-ISOLDE isotope separator facility. Neutron deficient mercury isotopes were produced by proton-induced spallation of lead nuclei: a molten lead target was bombarded with protons from the CERN Proton Synchrotron booster at an energy of $1.4\ \text{GeV}$. The proton beam intensity (1×10^{13} protons per pulse, up to $1\ \mu\text{A}$ averaged current) and bunched proton extraction was optimized to enhance the release of short-lived isotopes from the liquid target whilst reducing the likelihood of a mechanical failure due to shockwaves generated by the proton impact^{24,25}. A temperature-controlled spiral chimney enables effusion of the mercury vapor towards the ion source cavity whilst ensuring the required suppression, by condensation, of less volatile species such as lead. The mercury atoms effuse into the anode cavity of a FEBIAD-type ion source, referred to as the VADLIS (Versatile Arc Discharge and Laser Ion Source)³. For this experiment the laser beams from the Res-

onance Ionization Laser Ion Source (RILIS)²⁶ setup were directed into the FEBIAD anode cavity through the $1.5\ \text{mm}$ ion extraction aperture. The lasers were tuned to the 3-step resonance ionization scheme for mercury (Figure 1a inset), resulting in laser-ionization of mercury only. For the first time during on-line operation, the ion source was operated in the newly-established RILIS-mode³, whereby the anode voltage is reduced to several volts, whilst the cathode heating and ion source magnetic field strength are optimized for laser-ion survival and extraction. In this case, no electron impact ionization occurs, and the laser-ionized Hg^+ beam purity is maximized. The ion source efficiency remains similar to that of standard FEBIAD operation with electron-impact ionization (estimated to be several percent). Mercury ions leaving the anode exit aperture are subject to the $30\ \text{kV}$ potential difference between the ion source aperture and the grounded extraction electrode. The resulting mono-energetic beam of mercury ions was then passed through a magnetic dipole mass separator, which has sufficient resolution for the selection of the atomic mass of interest for transmission to the detection system, but is not capable of separating isobars.

Laser spectroscopy and signal identification

To extend our reach beyond the range of previously studied mercury isotopes, it was necessary to use the most sensitive laser spectroscopy method available at ISOLDE: in-source resonance ionization spectroscopy. As the name implies, this approach makes use of the initial ion creation process for performing laser spectroscopy, by wavelength-tuning of one of the lasers used in the resonance ionization scheme, which directly influences the ionization efficiency of the element of interest. For this work the $254\ \text{nm}$ first-step transition ($6s^2\ ^1\text{S}_0 \rightarrow 6d6p\ ^3\text{P}_1$)[◦] of the ionization scheme was used for spectroscopy since its sensitivity to nuclear variations results in a well-resolved hyperfine structure even in the high-temperature ($\approx 1800\ ^\circ\text{C}$) Doppler-broadening dominated environment of the ion source cavity. The $254\ \text{nm}$ light required was generated by third-harmonic generation from the output of a Ti:Sapphire laser, operating in the dual etalon scanning mode (fundamental linewidth of $0.8\ \text{GHz}$)²⁷. While operating the ion source in RILIS-mode, this laser was scanned in a stepwise manner and at each frequency step the ion rate was recorded using the most suitable of the three detection methods available. Depending on the intensity, purity, lifetime or decay mode of the beam at a chosen mass, the ion beam was directed to one of the three detection systems depicted in Figure 1a: The ISOLDE Faraday cup, for ion current measurement above $100\ \text{fA}$; the ISOLTRAP MR-ToF MS, for isobar-separated ion counting of longer lived isotopes with a resolving power of $M/\Delta M \approx 300000$ ^{28,29}; or the Windmill decay spectroscopy setup (for efficient detection of alpha-decaying isotopes). Laser scans were obtained for all $^{177}\text{--}^{186}\text{Hg}$ ground states. The isotope shifts were determined relative to ^{198}Hg . A total of $17\ ^{198}\text{Hg}$ reference scans were recorded periodically during the experiment.

Monte Carlo Shell Model Calculations

The Monte Carlo Shell Model (MCSM) calculation is one of the most advanced computational methods that can be applied for nuclear many-body systems. This work represents the largest ever MCSM calculations, performed on massively parallel supercomputers, including the K computer in Kobe, Japan. Exploiting the advantages of quantum Monte-Carlo, variational and matrix-diagonalization methods, this approach circumvents the diagonalization of a $> 2 \times 10^{42}$ -dimensional Hamiltonian matrix. Using the doubly-magic ^{132}Sn nucleus as an inert core, 30 protons and up to 24 neutrons were left to actively interact in an exceptionally large model space as compared to conventional CI calculations. Single-particle energies were set to be consistent with single-particle properties of ^{132}Sn and ^{208}Pb with smooth changes as functions of Z and N . All nucleons interact through effective NN interactions adopted from the frequently-used ones provided by^{22,23}. The Hamiltonian was thus fixed and kept throughout this study. The model space in which the nucleons interact are 11 proton orbits from $1g_{7/2}$ up to $1i_{13/2}$ and 13 neutron orbits from $1h_{9/2}$ up to $1j_{15/2}$. Calculations were performed utilizing effective charges for protons and neutrons, with their actual values being 1.6e and 0.6e, respectively and a spin quenching factor of 0.9. The MCSM eigenstate is represented as a superposition of MCSM basis vectors with the appropriate projection onto spin and parity. Each MCSM basis vector is a Slater determinant formed by mixed single-particle states, where the mixing amplitudes are optimized by quantum Monte-Carlo and variational methods. This Slater determinant has intrinsic quadrupole moments, which can be expressed in terms of a set of β_2 and γ deformation parameters. Using the effective charges and spin quenching factor, we reproduce the electric quadrupole moments of $^{177,179}\text{Hg}$ ³⁰ and the magnetic dipole moments of the odd-mass mercury isotopes (see Figure 2b). The combined effect of the monopole interaction, whose effect depends linearly on the orbital occupancy numbers, and the quadrupole interaction, that contains a quadratic dependence with a maximum when an orbital is half-filled, causes the observed shape staggering. Specifically the strong attractive monopole matrix element between proton $h_{9/2}$ and neutron $i_{13/2}$ orbitals (-0.35 MeV) compared to an average value of -0.2 MeV for the other orbitals considered, localizes the observed phenomena around $N = 103$ ¹⁴. This is due to the similarity in radial wave functions and the coherent sizable effect of the tensor force²³.

Using the effective charges, the quadrupole moments for each MCSM basis vector can be obtained. These can be converted into the standard ellipsoidal nuclear shape parameters β_2 (deformation strength) and γ (deformation shape)³¹. One of the strengths of the MCSM is the ability to link the basis vectors onto the Potential Energy Surface (PES) deduced using the same NN interaction. The T-plot^{14,32} was introduced in order to indicate the individual MCSM basis vectors using β_2 and γ as partial coordinates on the PES.

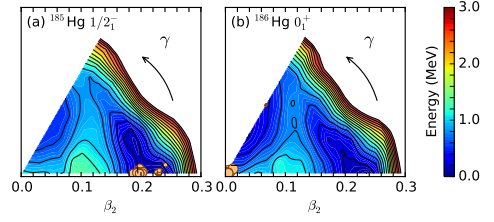


Figure 3. T-plot examples for two different states in $^{185,186}\text{Hg}$, with large and small deformation respectively. The corresponding β_2 and γ shape parameters of the main contributing MCSM basis vectors are denoted by the yellow circles.

Figure 3 displays example T-plots for $^{185,186}\text{Hg}$. The importance of each MCSM basis vector is evaluated in terms of the probability amplitude in the eigenstate of interest, and is shown by the size of the circle: larger circles representing major MCSM basis vectors, whilst smaller circles imply a less relevant contribution to this particular eigenstate. In the T-plot, clustering of the circles is observed for spherical (^{186}Hg) and prolate shapes (^{185}Hg).

In order to compare the $\delta\langle r^2 \rangle$ from the isotope shift measurements to the β_2 deduced from the MCSM calculations, the following standard procedure was used:

$$\langle r^2 \rangle = \langle r^2 \rangle_{\text{DM}} \left(1 + \frac{5}{4\pi} \langle \beta_2^2 \rangle \right) \quad (1)$$

$$\delta\langle r^2 \rangle_{A-186} = \langle r^2 \rangle - \langle r^2 \rangle_{186} \quad (2)$$

where $\langle r^2 \rangle_{\text{DM}}$ refers to the droplet model mean square charge radius for which the most-used parametrization of Berdichevsky and Tondeur³³ is adopted. Following this procedure, the influence of γ is negligible.

The calculated $\delta\langle r^2 \rangle$, shown in Fig. 2a, are indicated by shaded boxes. The height of the box is determined by the distribution of the circles in the T-plot with respect to β_2 and represents the spread of the quantum fluctuations of the MCSM eigenstates.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

1. Bonn, J., Huber, G., Kluge, H.-J., Kugler, L. & Otten, E. Sudden change in the nuclear charge distribution of very light mercury isotopes. *Phys. Lett. B* **38**, 308–311 (1972). URL <https://doi.org/10.1016/2F0370-2693%2872%2990253-5>. DOI 10.1016/0370-2693(72)90253-5.

2. Kühl, T. *et al.* Nuclear shape staggering in very neutron-deficient Hg isotopes detected by laser spectroscopy. *Phys. Rev. Lett.* **39**, 180–183 (1977). URL <https://doi.org/10.1103/PhysRevLett.39.180>. DOI 10.1103/PhysRevLett.39.180.
3. Day Goodacre, T. *et al.* Blurring the boundaries between ion sources: The application of the RILIS inside a FEBIAD-type ion source at ISOLDE. *Nucl. Instruments Methods Phys. Res. Sect. B: Beam Interactions with Mater. Atoms* **376**, 39–45 (2016). URL <https://doi.org/10.1016/j.nimb.2016.03.005>. DOI 10.1016/j.nimb.2016.03.005.
4. Ulm, G. *et al.* Isotope shift of ^{182}Hg and an update of nuclear moments and charge radii in the isotope range ^{181}Hg – ^{206}Hg . *Zeitschrift für Physik A At. Nucl.* **325**, 247–259 (1986). URL <https://doi.org/10.1007/BFb01294605>. DOI 10.1007/BFb01294605.
5. Witte, H. D. *et al.* Nuclear charge radii of neutron-deficient lead isotopes beyond $n = 104$ midshell investigated by in-source laser spectroscopy. *Phys. Rev. Lett.* **98** (2007). URL <https://doi.org/10.1103/PhysRevLett.98.112502>. DOI 10.1103/PhysRevLett.98.112502.
6. Andreyev, A. N. *et al.* A triplet of differently shaped spin-zero states in the atomic nucleus ^{186}Pb . *Nat.* **405**, 430–433 (2000). URL <https://doi.org/10.1038/2F35013012>. DOI 10.1038/35013012.
7. Wood, J. L. & Heyde, K. A focus on shape coexistence in nuclei. *J. Phys. G: Nucl. Part. Phys.* **43**, 020402 (2016). URL <https://doi.org/10.1088/2F0954-3899/43/2/020402>. DOI 10.1088/0954-3899/43/2/020402.
8. Campbell, P., Moore, I. & Pearson, M. Laser spectroscopy for nuclear structure physics. *Prog. Part. Nucl. Phys.* **86**, 127–180 (2016). URL <https://doi.org/10.1016/j.pnpnp.2015.09.003>. DOI 10.1016/j.pnpnp.2015.09.003.
9. Heyde, K. & Wood, J. L. Shape coexistence in atomic nuclei. *Rev. Mod. Phys.* **83**, 1467–1521 (2011). URL <https://doi.org/10.1103/RevModPhys.83.1467>. DOI 10.1103/RevModPhys.83.1467.
10. Wrzosek-Lipska, K. & Gaffney, L. P. Unique and complementary information on shape coexistence in the neutron-deficient Pb region derived from coulomb excitation. *J. Phys. G: Nucl. Part. Phys.* **43**, 024012 (2016). URL <https://doi.org/10.1088/2F0954-3899/43/2/024012>. DOI 10.1088/0954-3899/43/2/024012.
11. Julin, R., Grahn, T., Pakarinen, J. & Rahkila, P. In-beam spectroscopic studies of shape coexistence and collectivity in the neutron-deficient $z \approx 82$ nuclei. *J. Phys. G: Nucl. Part. Phys.* **43**, 024004 (2016). URL <https://doi.org/10.1088/2F0954-3899/43/2/024004>. DOI 10.1088/0954-3899/43/2/024004.
12. Borge, M. J. G. & Blaum, K. Focus on Exotic Beams at ISOLDE: A Laboratory Portrait. *J. Phys. G: Nucl. Part. Phys.* **45**, 010301 (2017). URL <https://doi.org/10.1088/2F1361-6471/45/1/010301>. DOI 10.1088/1361-6471/aa990f.
13. Otsuka, T., Honma, M., Mizusaki, T., Shimizu, N. & Utsuno, Y. Monte carlo shell model for atomic nuclei. *Prog. Part. Nucl. Phys.* **47**, 319 – 400 (2001). URL <http://www.sciencedirect.com/science/article/pii/S0146641001001570>. DOI [https://doi.org/10.1016/S0146-6410\(01\)00157-0](https://doi.org/10.1016/S0146-6410(01)00157-0).
14. Otsuka, T. & Tsunoda, Y. The role of shell evolution in shape coexistence. *J. Phys. G: Nucl. Part. Phys.* **43**, 024009 (2016). URL <https://doi.org/10.1088/2F0954-3899/43/2/024009>. DOI 10.1088/0954-3899/43/2/024009.
15. Sachdev, S. *Quantum Phase Transitions - Second Edition* (Cambridge University Press, 2011).
16. Carr, L. D. *Understanding Quantum Phase Transitions* (Crc Press Inc., 2010).
17. Togashi, T., Tsunoda, Y., Otsuka, T. & Shimizu, N. Quantum phase transition in the shape of Zr isotopes. *Phys. Rev. Lett.* **117**, 172502 (2016). URL <https://link.aps.org/doi/10.1103/PhysRevLett.117.172502>. DOI 10.1103/PhysRevLett.117.172502.
18. Bree, N. *et al.* Shape coexistence in the neutron-deficient even-even $^{182-188}\text{Hg}$ isotopes studied via coulomb excitation. *Phys. Rev. Lett.* **112** (2014). URL <https://doi.org/10.1103/PhysRevLett.112.162701>. DOI 10.1103/PhysRevLett.112.162701.
19. Marsh, B. *et al.* New developments of the in-source spectroscopy method at RILIS/ISOLDE. *Nucl. Instruments Methods Phys. Res. Sect. B: Beam Interactions with Mater. Atoms* **317**, 550–556 (2013). URL <https://doi.org/10.1016/j.nimb.2013.07.070>. DOI 10.1016/j.nimb.2013.07.070.
20. Day Goodacre, T. *et al.* RILIS-ionized mercury and tellurium beams at ISOLDE/CERN. *Hyperfine Interactions* **238** (2017). URL <https://doi.org/10.1007/978-94-007-1398-6>. DOI 10.1007/978-94-007-1398-6.
21. Wolf, R. *et al.* ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer. *Int. J. Mass Spectrom.* **349-350**, 123–133 (2013). URL <https://doi.org/10.1016/j.ijms.2013.03.020>. DOI 10.1016/j.ijms.2013.03.020.
22. Brown, B. A. Double-octupole states in ^{208}Pb . *Phys. Rev. Lett.* **85**, 5300–5303 (2000). URL <https://doi.org/10.1103/PhysRevLett.85.5300>.

- org/10.1103/PhysRevLett.85.5300. DOI 10.1103/PhysRevLett.85.5300.
23. Otsuka, T. *et al.* Novel features of nuclear forces and shell evolution in exotic nuclei. *Phys. Rev. Lett.* **104** (2010). URL <https://doi.org/10.1103/PhysRevLett.104.012501>. DOI 10.1103/PhysRevLett.104.012501.
 24. Lettry, J. *et al.* Release from ISOLDE molten metal targets under pulsed proton beam conditions. *Nucl. Instruments Methods Phys. Res. Sect. B: Beam Interactions with Mater. Atoms* **126**, 170–175 (1997). URL <https://doi.org/10.1016/2Fs0168-583x%2896%2901088-9>. DOI 10.1016/S0168-583X(96)01088-9.
 25. Lettry, J. *et al.* Effects of thermal shocks on the release of radioisotopes and on molten metal target vessels. *Nucl. Instruments Methods Phys. Res. Sect. B: Beam Interactions with Mater. Atoms* **204**, 251–256 (2003). URL <https://doi.org/10.1016/2Fs0168-583x%2802%2901919-5>. DOI 10.1016/S0168-583X(02)01919-5.
 26. Fedosseev, V. *et al.* Ion beam production and study of radioactive isotopes with the laser ion source at ISOLDE. *J. Phys. G: Nucl. Part. Phys.* **44**, 084006 (2017). URL <https://doi.org/10.1088/2F1361-6471%2Faa78e0>. DOI 10.1088/1361-6471/aa78e0.
 27. Rothe, S. *et al.* Narrow linewidth operation of the RILIS titanium: Sapphire laser at ISOLDE/CERN. *Nucl. Instruments Methods Phys. Res. Sect. B: Beam Interactions with Mater. Atoms* **317**, 561–564 (2013). URL <https://doi.org/10.1016/2Fj.nimb.2013.08.058>. DOI 10.1016/j.nimb.2013.08.058.
 28. Wolf, R. N., Erritt, M., Marx, G. & Schweikhard, L. A multi-reflection time-of-flight mass separator for isobaric purification of radioactive ion beams. *Hyperfine Interactions* **199**, 115–122 (2011). URL <https://doi.org/10.1007/2Fs10751-011-0306-8>. DOI 10.1007/s10751-011-0306-8.
 29. Wienholtz, F. *et al.* Towards ultra high-resolution multi-reflection time-of-flight mass spectrometry at ISOLTRAP. *Phys. Scripta* **T166**, 014068 (2015). URL <https://doi.org/10.1088/2F0031-8949%2F2015%2Ft166%2F014068>. DOI 10.1088/0031-8949/2015/t166/014068.
 30. Sels, S. *In-source resonance-ionization spectroscopy of neutron-deficient $^{177-185}\text{Hg}$ isotopes* (In preparation, 2018).
 31. Bohr, A. & Mottelson, B. R. *Nuclear Structure* (World Scientific Publishing Company, 1998). URL <https://doi.org/10.1142/2F3530-vol2>.
 32. Tsunoda, Y., Otsuka, T., Shimizu, N., Honma, M. & Utsuno, Y. Novel shape evolution in exotic ni isotopes and configuration-dependent shell structure. *Phys. Rev. C* **89**, 031301 (2014). URL <https://link.aps.org/doi/10.1103/PhysRevC.89.031301>. DOI 10.1103/PhysRevC.89.031301.
 33. Berdichevsky, D. & Tondeur, F. Nuclear core densities isotope shifts, and the parametrization of the droplet model. *Zeitschrift für Physik A Atoms Nucl.* **322**, 141–147 (1985). URL <https://doi.org/10.1007/2Fb01412027>. DOI 10.1007/bf01412027.
 34. Day Goodacre, T. *Developments of the ISOLDE RILIS for radioactive ion beam production and the results of their application in the study of exotic mercury isotopes*. Ph.D. thesis, School of Physics and Astronomy, The University of Manchester (2016).
 35. Sels, S. *Laser spectroscopy of mercury - Return towards sphericity*. Ph.D. thesis, Instituut voor Kern en Stralingsfysica, K.U. Leuven, B-3001 Leuven, Belgium (2018).
 36. Reimann, R. J. & McDermott, M. N. Precision magnetic moment determinations for 43-min $^{199\text{m}}\text{Hg}$ and other isomers of mercury. *Phys. Rev. C* **7**, 2065–2079 (1973). URL <https://link.aps.org/doi/10.1103/PhysRevC.7.2065>. DOI 10.1103/PhysRevC.7.2065.
 37. Kohler, R. H. Detection of double resonance by frequency change: Application to ^{201}Hg . *Phys. Rev.* **121**, 1104–1111 (1961). URL <https://link.aps.org/doi/10.1103/PhysRev.121.1104>. DOI 10.1103/PhysRev.121.1104.

Supplementary Information

The isotope shifts and corresponding $\delta\langle r^2 \rangle$ values are provided in Table 1. A complete description of the data analysis and fitting can be found in the Doctoral theses of T. Day Goodacre³⁴ and S. Sels³⁵. The close agreement between the values extracted from this work with those of literature is a convincing validation of the data treatment.

Acknowledgements

This work has received funding through the following channels:

The Max-Planck-Society, IMPRS-PTFS; BMBF (German Federal Ministry for Education and Research) nos. 05P12HGCI1 and 05P15HGCI1; STFC Grants No. ST/L005727, ST/M006433, ST/M006434, ST/L002868/1, ST/L005794/1; Slovak Research and Development Agency, contract No. APVV-14-0524; European Unions Seventh Framework Programme for Research and Technological Development under Grant Agreement 267194 (COFUND) and 289191. FWO-Vlaanderen (Belgium), by GOA/2015/010 (BOF KU Leuven); the Inter-university Attraction Poles Programme initiated by the Belgian Science Policy Office (BriX network P7/12), by the European Commission within

Table 1. Summary of resulting mean-square charge differences ($\delta\langle r^2 \rangle^{A-198}$) and nuclear moments (μ , Q) and their comparison to literature.

Isotope	Spin I^π	$\delta\langle r^2 \rangle^{A-198}$ (this work) (fm ²)	$\delta\langle r^2 \rangle^{A-198}$ (lit.) (fm ²)	μ (this work) (μ_N)	μ (lit.) (μ_N)	Q_s (this work) (b)	Q_s (lit.) (b)
¹⁷⁷ Hg	(7/2 ⁻)	-1.067(8){78}	-	-1.027(53)	-	0.57(83)	-
¹⁷⁸ Hg	0 ⁺	-0.968(6){71}	-	-	-	-	-
¹⁷⁹ Hg	(7/2 ⁻)	-0.905(5){70}	-	-0.949(29)	-	0.77(28)	-
¹⁸⁰ Hg	0 ⁺	-0.808(5){60}	-	-	-	-	-
¹⁸¹ Hg	1/2 ⁻	-0.111(6){11}	-0.114(4){10}	0.510(9)	0.5071(7)	-	-
¹⁸² Hg	0 ⁺	-0.653(5){48}	-0.649(10){49}	-	-	-	-
¹⁸³ Hg	1/2 ⁻	-0.065(5){7}	-0.069(2){6}	0.516(11)	0.524(5)	-	-
¹⁸⁴ Hg	0 ⁺	-0.542(5){40}	-0.544(2){42}	-	-	-	-
¹⁸⁵ Hg	1/2 ⁻	-0.069(6){7}	-0.0764(6){63}	0.507(17)	0.509(4)	-	-
^{185m} Hg	13/2 ⁺	-0.543(4){40}	-0.543(2){42}	-1.009(12)	-1.017(9)	-0.15(41)	0.19(32)

Statistical errors are given in parenthesis. Systematic errors stemming from the indeterminacy of the F factor (7%, see⁴) and M_{SMS} are shown in curly brackets
Ref. for μ : $\mu(^{199}\text{Hg}^m) = -1.0147(8)$, $a(^{199}\text{Hg}^m) = -2298.3(2)$ ³⁶
Ref. for Q_s : $Q_s(^{201}\text{Hg}) = 0.387(6)^?$, $b(^{201}\text{Hg}) = -280.107(5)$ ³⁷

the Seventh Framework Programme through I3-ENSAR (contract no. RII3-CT-2010-262010) and by a grant from the European Research Council (ERC-2011-AdG-291561-HELIOS).

S.S acknowledges the Agency for Innovation by Science and Technology in Flanders (IWT).

L.P.G acknowledges FWO-Vlaanderen (Belgium) as an FWO Pegasus Marie Curie Fellow. A.W and K.Z acknowledge the Wolfgang-Gentner scholarship and the BMBF (German Federal Ministry for Education and Research) no. 05P120DCIA. This work was supported by JSPS and FWO under the Japan-Belgium Research Cooperative Program. The MCSM calculations were performed on the K computer at RIKEN AICS (hp160211, hp170230). This work was also supported in part by ‘Priority Issue on Post-K computer’(Elucidation of the Fundamental Laws and Evolution of the Universe) from MEXT and JICFuS.

ation; B.A, D.A, T.E.C, J.G.C, K.T.F, L.P.G, L.G, K.M.L, V.M, M.R, R.E.R, L.S, S.S, C.V.B, A.W, F.W, R.N.W set up and operated the detection and data acquisition systems; A.A, A.B, K.B, T.E.C, T.D.G, J.D, D.V.F, L.P.G, M.H, B.A.M, T.O, L.S, M.D.S, S.S, P.V.D, E.V, K.W contributed to the data analysis and interpretation; A.A, A.B, K.B, T.E.C, J.G.C, T.D.G, J.D, V.N.F, L.P.G, M.H, D.L, B.A.M, T.O, L.S, S.S, Y.T, P.V.D contributed to the manuscript preparation.

Author contributions statement

A.A, A.B, T.D.G, D.V.F, V.N.F, L.P.G, M.H, B.A.M, M.D.S, P.V.D conceived the experiment and prepared the proposal; T.D.G, D.V.F, V.N.F, B.A.M, Y.M.P, P.L.M, R.E.R, S.R, M.V carried out laser ionization scheme and/or ion source developments; A.A, K.B, T.E.C, V.N.F, M.H, S.K, B.A.M, L.S, P.V.D, K.Z supervised the participants; B.A, N.A.A, D.A, A.B, J.B, T.E.C, J.G.C, T.D.G, G.J.F, D.V.F, V.N.F, K.T.F, L.P.G, L.G, M.H, K.M.L, V.M, B.A.M, Y.M.P, P.L.M, R.E.R, S.S, P.S, C.V.B, P.V.D, M.V, E.V, A.W, F.W, A.Z participated in data taking; T.O, Y.T performed MCSM calculations; J.D, A.P performed DFT calculations; A.B, T.D.G, D.V.F, B.A.M, P.L.M, R.E.R, S.R, M.V took part in laser set up and oper-

Shape staggering of mid-shell mercury isotopes from in-source laser spectroscopy compared with Density Functional Theory and Monte Carlo Shell Model calculations

S. Sels,^{1,*} T. Day Goodacre,^{2,3} B. A. Marsh,³ A. Pastore,⁴ W. Ryssens,⁵ Y. Tsunoda,⁶ N. Althubiti,² B. Andel,⁷ A. N. Andreyev,^{4,8} D. Atanasov,⁹ A. E. Barzakh,¹⁰ M. Bender,⁵ J. Billowes,² K. Blaum,⁹ T. E. Cocolios,¹ J. G. Cubiss,⁴ J. Dobaczewski,^{4,11} G. Farooq-Smith,¹ D. V. Fedorov,¹⁰ V. N. Fedosseev,³ K. T. Flanagan,² L. P. Gaffney,^{12,1} L. Ghys,^{13,1} P.-H. Heenen,¹⁴ M. Huyse,¹ S. Kreim,⁹ D. Lunney,¹⁵ K. M. Lynch,³ V. Manea,⁹ Y. Martinez Palenzuela,¹ T. M. Medonca,³ P. L. Molkanov,¹⁰ T. Otsuka,^{6,16,1} J. P. Ramos,^{3,17} R. E. Rossel,^{3,18} S. Rothe,³ L. Schweikhard,¹⁹ M. D. Seliverstov,¹⁰ P. Spagnoletti,¹² C. Van Beveren,¹ P. Van Duppen,¹ M. Veinhard,³ E. Verstraelen,¹ A. Welker,²⁰ K. Wendt,¹⁸ F. Wienholtz,¹⁹ R.N. Wolf,⁹ and A. Zadornaya¹

¹*KU Leuven, Instituut voor Kern- en Stralingsfysica, B-3001 Leuven, Belgium*

²*School of Physics and Astronomy, The University of Manchester, Manchester M13 9PL, UK*

³*CERN, CH-1211 Geneve 23, Switzerland*

⁴*Department of Physics, University of York, York YO10 5DD, UK*

⁵*IPNL, Université de Lyon, Université Lyon 1, CNRS/IN2P3, F-69622 Villeurbanne, France*

⁶*Department of Physics, The University of Tokyo, Bunkyo-ku, 113-0033 Tokyo, Japan*

⁷*Department of Nuclear Physics and Biophysics,*

Comenius University in Bratislava, 84248 Bratislava, Slovakia

⁸*Advanced Science Research Center, Japan Atomic Energy Agency (JAEA), Tokai-mura, Japan*

⁹*Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany*

¹⁰*Petersburg Nuclear Physics Institute, NRC Kurchatov Institute, Gatchina 188300, Russia*

¹¹*Institute of Theoretical Physics, Faculty of Physics,*

University of Warsaw, ul. Pasteura 5, PL-02-093 Warsaw, Poland

¹²*School of Engineering and Computing, University of the West of Scotland, Paisley, PA1 2BE, UK*

¹³*Belgian Nuclear Research Center SCK•CEN, Boeretang 200, B-2400 Mol, Belgium*

¹⁴*PNTPM, CP229, Université Libre de Bruxelles, B-1050 Bruxelles, Belgium*

¹⁵*CSNSM-IN2P3-CNRS, Université Paris-Sud, 91406 Orsay, France*

¹⁶*National Superconducting Cyclotron Laboratory,*

Michigan State University, East Lansing, MI 48824, USA

¹⁷*Laboratory of Powder Technology, École polytechnique fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland*

¹⁸*Insitut für Physik, Johannes Gutenberg-Universität, 55122 Mainz, Germany*

¹⁹*Institut für Physik, Universität Greifswald, 17487 Greifswald, Germany*

²⁰*Technische Universität Dresden, 01069 Dresden, Germany*

(Dated: September 12, 2018)

Neutron-deficient ^{177–185}Hg isotopes were studied using in-source laser resonance-ionization spectroscopy at the CERN-ISOLDE radioactive ion-beam facility, in an experiment combining different detection methods tailored to the studied isotopes. These include either α -decay tagging or Multi-reflection Time-of-Flight gating to identify the isotopes of interest. The endpoint of the odd-even nuclear shape staggering in mercury was observed directly by measuring for the first time the isotope shifts and hyperfine structures of ^{177–180}Hg. Changes in the mean-square charge radii for all mentioned isotopes, magnetic dipole and electric quadrupole moments of the odd-*A* isotopes and arguments in favor of $I = 7/2$ spin assignment for ^{177,179}Hg were deduced. Experimental results are compared with Density Functional Theory (DFT) and Monte-Carlo Shell Model (MCSM) calculations. DFT calculations with several Skyrme parameterizations predict a large jump in the charge radius around the neutron $N = 104$ mid shell, with an odd-even staggering pattern related to the coexistence of nearly-degenerate oblate and prolate minima. This near-degeneracy is highly sensitive to many aspects of the effective interaction, a fact that renders perfect agreement with experiment out of reach for current functionals. Despite this inherent difficulty, the SLy5s1 and a modified UNEDF1^{SO} parameterization predict a qualitatively correct staggering that is off by two neutron numbers. MCSM calculations of states with the experimental spins and parities show good agreement for both electromagnetic moments and the observed charge radii. A clear mechanism for the origin of shape staggering within this context is identified: a substantial change in occupancy of the proton $\pi h_{9/2}$ and neutron $\nu i_{13/2}$ orbitals.

I. Introduction

More than four decades ago, an unexpected large difference in the mean-square charge radius between ¹⁸⁷Hg and ¹⁸⁵Hg was observed by measuring the isotope shift in

* Simon.Sels@kuleuven.be

a Radiation Detection of Optical Pumping (RADOP) experiment performed at ISOLDE [1, 2]. Similar to ^{185}Hg , the $^{181,183}\text{Hg}$ isotopes were found to exhibit a large isotope shift from their even-mass neighbours $^{182,184,186}\text{Hg}$ [3, 4]. Ever since these measurements, the observed pattern became known as ‘shape staggering’. Studying the levels at low excitation energy in more detail, different shapes were identified in close vicinity to the ground state and the mercury isotopes are now one of the most illustrative examples of shape coexistence [5]. The experimental findings sparked extensive interest in studying this region of the nuclear chart from both experimental and theoretical points of view [5]. The large radius staggering was interpreted as transitions between weakly-deformed, oblate ground states and strongly-deformed, prolate ground states [6].

The isotopic chain of mercury has since been studied with a multitude of complementary techniques: Coulomb excitation [7–9], in-beam gamma-ray spectroscopy with recoil-decay tagging [10–13], mass measurements [14] and α/β -decay spectroscopy [15–21]. However, isotope shift and hyperfine-structure measurements had only been extended down to ^{181}Hg [2, 4]. While the ground-state deformation has been indirectly inferred for neutron-deficient mercury isotopes from in-beam recoil-decay tagging measurements, hinting towards less-deformed shapes for $A < 180$ [10–13], this had not been confirmed by a direct ground-state isotope-shift measurement. The missing mean-square charge-radii data for the lighter mercury isotopes left the key question of where the shape staggering ends.

In order to address this key question, a measurement campaign was undertaken at the radioactive ion-beam facility ISOLDE [22] performing in-source laser resonance-ionization spectroscopy of 15 mercury isotopes, ranging from the neutron-deficient to the neutron-rich side ($^{177}–^{185}, ^{198}, ^{202}, ^{203}, ^{206}–^{208}\text{Hg}$) with the goal of measuring their isotope/isomer shifts (IS) and hyperfine structures (HFS). The large isotopic span was made possible by using the Resonance Ionization Laser Ion Source (RILIS) [23] in a novel target-ion source combination [24], together with three different ion-counting techniques tailored to the isotope under investigation [25]: α -decay spectroscopy for short-lived isotopes with small production rates (down to 0.1 ion/s) using a ‘Windmill’-type implantation station (WM), Multi-Reflection Time-of-Flight mass spectrometer/separators (MR-ToF MS) for high-resolution, single-ion counting and Faraday Cup (FC) ion-current measurements for high-intensity (>1 pA) mercury beams. This paper is an in-depth follow-up article of [26] on the neutron-deficient isotopes $^{177}–^{185}\text{Hg}$. A dedicated paper will provide a detailed discussion of the neutron-rich isotopes [27] also measured in the same experimental campaign.

II. Experimental Technique

A. Mercury ion beam production

Mercury isotopes were produced at the CERN-ISOLDE facility [22] via spallation reactions induced by a 1.4-GeV proton beam from the PS-Booster synchrotron impinging upon a molten-lead target. The neutral reaction products effused from the heated target via the transfer line ($\approx 700^\circ\text{C}$ target and $\approx 400^\circ\text{C}$ transfer line heating) into the VADLIS cavity, which was operated in RILIS mode [24] (Fig. 1). In this mode, lasers are used to resonantly ionize the isotopes of interest. The photo-ions were extracted and accelerated by a 30 kV potential difference, mass separated by ISOLDE’s General Purpose Separator (GPS) dipole bending magnet before being sent to one of three measurement devices (FC/WM/MR-ToF MS; see Fig. 1). The choice of a molten-lead target was based on results obtained from a preparatory experiment in similar conditions at ISOLDE, where the mercury production of a molten-lead was compared with a UC_x target (Fig. 2). While the production rates of the lightest mercury isotopes were of a similar order of magnitude for both cases, the use of a molten-lead target significantly reduces the isobaric contamination of surface-ionised contaminants. This was especially important for the heavy mass region discussed in [27]. Further-

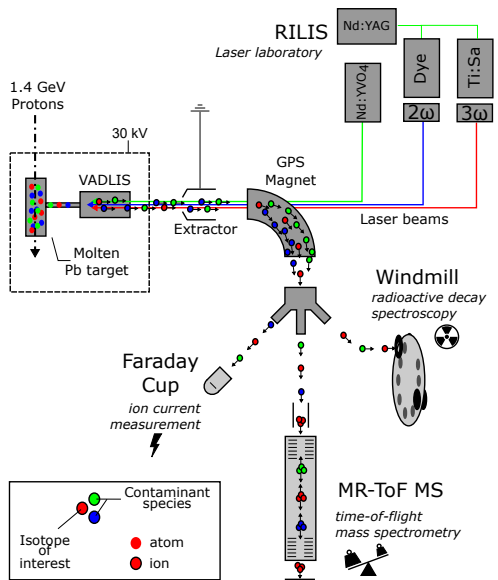


FIG. 1. Schematic of the experimental setup in the ISOLDE facility at CERN. See text for details.

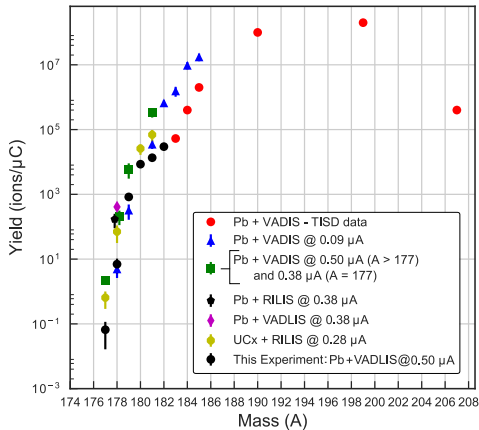


FIG. 2. Mercury production yield data for different target-ion source configurations: Versatile Arc Discharge Laser Ion Source (VADLIS) or Resonance Ionization Laser Ion Source (RILIS) with lead or uranium-carbide target material for different proton-beam currents.

more, the production rate of the heavy mercury isotopes was significantly higher for the molten-lead target.

Resonance ionization of the mercury isotopes was accomplished using a 3-step ionization scheme (Fig. 3) with a measured ionization efficiency of 6% [28]. Laser spectroscopy was performed on the 253.65-nm $6s^2 \ ^1S_0 \rightarrow 6s6p^3 \ ^3P_1$ transition. Well-resolved HFS spectra were obtained by scanning the frequency-tripled wavelength of the Ti:Sapphire laser with a bandwidth of ≈ 1.5 -GHz FWHM after tripling (labelled 3ω in Fig. 1). At the second step (313 nm) the frequency-doubled radiation of the Sirah Credo dye laser was used. The third step was a non-resonant 532-nm transition driven by a Nd:YVO₄ laser.

B. Isotope identification and counting

1. Ion-current measurement with a Faraday cup

A Faraday cup installed downstream the GPS magnet was used to measure the extracted ion current of sufficiently intense (>1 pA) beams of the longer-lived mercury isotopes. In this experiment, the reference isotope for IS measurements, stable ^{198}Hg , as well as radioactive $^{202,203}\text{Hg}$ were probed using this technique. In the case of ^{198}Hg , contamination of the neighboring ^{197}Hg ground and isomeric states was present in the HFS. To prove consistency of the IS measurements for different techniques, all isotopes measured using the FC were also

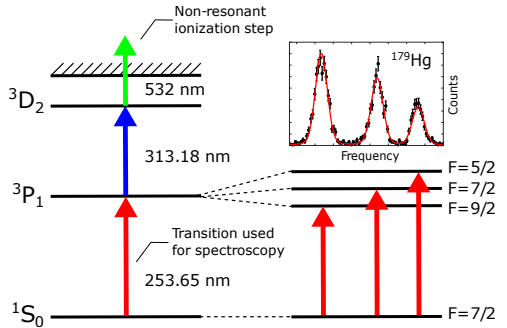


FIG. 3. Ionization scheme for mercury developed in [28], using the same transition for spectroscopy as in [1] and [29]. The right-hand side shows the splitting of the 3P_1 level states with different total angular momentum $F = I + J$ for an $I = 7/2$ nuclear spin with a corresponding exemplary HFS spectrum of ^{179}Hg . Energies and level splitting are not displayed to scale.

measured with the MR-ToF MS technique, with which it was possible to suppress the unwanted isotopic and isobaric contaminant species.

2. IS and HFS measurements with the Windmill setup

The Windmill detection setup [30, 31], consists of a vacuum chamber holding a rotatable wheel that houses 10 thin carbon foils up to 12-mm diameter ($20 \mu\text{g}/\text{cm}^2$ thickness) [32]. Surrounding these foils are two pairs of silicon detectors. The first pair is positioned around the carbon foil in which the beam is implanted. This pair consists of an annular (Ortec, TC-025-450-300-S, 6 mm hole, 450mm^2 active area) and a full surface barrier detector (Ortec, TB-020-300-500, 300mm^2 active area). The beam is implanted through the central hole of the annular detector. After implantation, the wheel rotates and a fresh foil is placed in view of the beam. The foil that was irradiated is moved towards the so-called ‘decay position’ between a second pair of silicon detectors. The two silicon detectors (Canberra, PD 300-15-300 RM, 300mm^2 active area) were used to study longer-lived radioactive isotopes that were implanted directly or the daughter products of previously implanted nuclei. The total α -particle detection efficiency at the implantation site was 34%, with a detector resolution of 35-keV FWHM. Two germanium detectors were additionally positioned outside the chamber of the WM setup, in order to measure γ - and X-ray radiation emitted from the implantation site.

The α -particle energy spectra obtained at the implantation point for different mass-separator settings are shown in Fig. 4. By using the α -particle energies to identify short-lived isotopes and associating the count rates with the wavenumber of the laser targeting the spectroscopic transition, it is possible to produce nearly-background-free HFS when the α particles coming from the decay of the beam contaminants have sufficiently differing energies. This is for instance shown in the α -energy spectrum collected with $A = 177$ mass-separator setting where contaminants from $^{178-185}\text{Hg}$ were present in the beam (Fig. 5). Here, clean HFS of 6 mercury isotopes can be observed in a single scan simply by gating on different α peaks. The efficiency and selectivity of this method allows IS and HFS measurements of isotopes with very small production rates such as ^{177}Hg which was delivered at a rate of ≈ 0.1 ions s^{-1} .

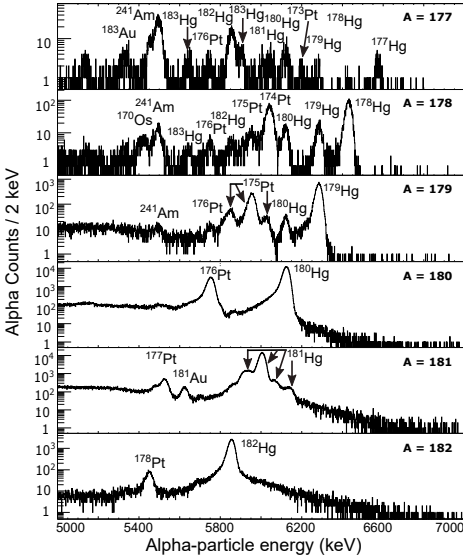


FIG. 4. α -particle energy spectra obtained at implantation site when the GPS was set to masses in the range of $A = 177-182$. Because of the limited mass resolution and difference in production, beam contaminants of heavier mercury isotopes are observed in the lighter-mass spectra.

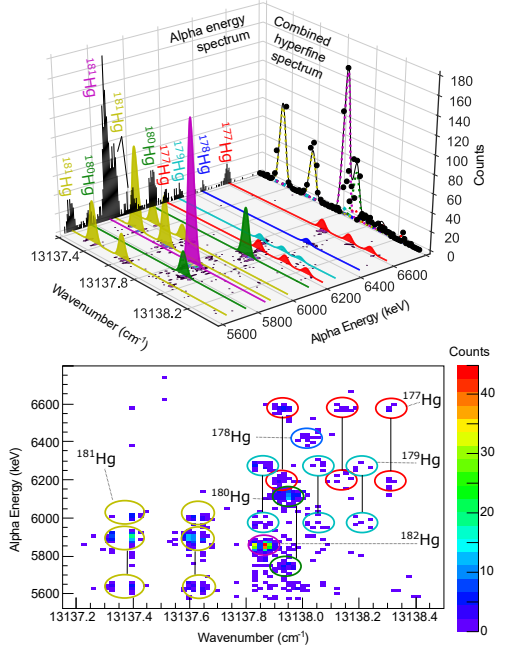


FIG. 5. Top: 3D-spectrum showing counts as function of α -decay energy and laser frequency before tripling (in wavenumber). The spectrum is obtained from a single laser scan measured at the Windmill implantation site when the GPS mass separator was set to $A = 177$. Because of the limited mass resolution and approximately four orders of magnitude larger production rate of ^{182}Hg in comparison to ^{177}Hg , beam contaminants are visible up to $A = 182$. The HFS for each contaminant is shown by the colored lines along the Z-axis, where different colors represent different mercury isotopes. When projecting on the α energy or laser-frequency axis, the total α -decay energy spectrum and combined HFS for all isotopes appear respectively. Bottom: The projection of the top plot on the α -decay energy and laser-frequency plane shows regions of counts (indicated by circles) related to the isotopes decay and HFS. Lines drawn between circles indicate the same HFS peaks visible in daughter products of the original decay-ing isotopes.

3. IS and HFS measurements with Multi-Reflection-Time-Of-Flight technique

For the measurement of $^{183-185,198,202,203,206-208}\text{Hg}$, ISOLTRAP's MR-ToF MS [33] was used for mass separation and ion detection. This device, extensively discussed in [34], consists of two 160 mm-long, 6-fold electrostatic mirrors surrounded by shielding electrodes.

First, the ion beam from ISOLDE is injected into a radio-frequency quadrupole cooler-buncher (RFQCB) [35]. Ion bunches are stopped and thermalized in this helium-filled RFQCB before they are injected into the MR-ToF MS with a typical energy spread of 60 eV and bunch width of 60 ns [33, 36]. Here, the ion bunches are trapped by reducing their kinetic energy and switching of the in-trap lift voltage. In the trapping cavity, they undergo multiple round-trips between the electrostatic mirrors, where the time-of-flight is dependent on the mass and the charge state. This causes a separation in time for different isobaric species in each bunch. The ion bunches are then ejected from the MR-ToF MS by switching the in-trap lift voltage [33]. The arrival time of the ejected, mass-separated ion bunches was measured by employing a MagnetTOFTM secondary electron multiplier ion detector (DM291, ETP, Ermington, Australia). An MR-TOF mass separator is able to reach resolving powers of $R = \Delta m/m = 10^5$ within a few ten milliseconds [33]. The procedure for employing the MR-ToF MS in a laser-spectroscopy experiment was previously discussed in [36]. Examples of different time-of-flight spectra are shown in Fig. 6.

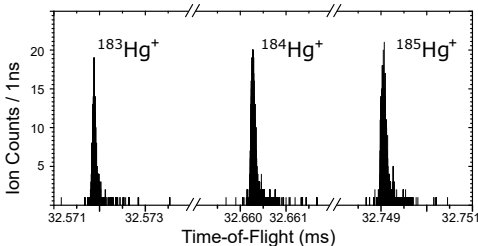


FIG. 6. Time-of-Flight spectra obtained for $^{183-185}\text{Hg}$, when the GPS mass separator was set to $A = 183$, $A = 184$, $A = 185$ respectively, after 1000 revolutions in the MR-ToF MS at which a mass resolving power of about 1.2×10^5 was reached. At this resolving power, all isotopic and isobaric contaminants are cleared from the spectra.

C. Recording of HFS

During the experiment, the wavelength of the laser targeting the spectroscopic transition was scanned in a step-wise manner and a parameter proportional to the number of detected photo-ions was recorded as a func-

tion of the scanned laser frequency. The data taking was synchronized with ISOLDE's super-cycle structure of proton pulses provided by the Proton Synchrotron Booster. Depending on the production rate of the isotope under investigation, between 1 and 5 full super-cycles of measurement were taken for each laser frequency step. Data obtained with the WM setup were recorded with an event-by-event data structure. The data were analyzed off-line with the ROOT software package [37], where the deadtime-corrected integral of energy-gated alpha counts for each laser frequency resulted in the hyperfine spectra. A similar analysis was performed for the MR-ToF MS data where isotope counting was not based on energy of an emitted particle but on ion arrival time at the ion detector. For measurements with the Faraday cup, the integrated ion current for each laser step was combined with the recorded laser frequency to create the HFS. An overview of the measured HFS is given in Fig. 7.

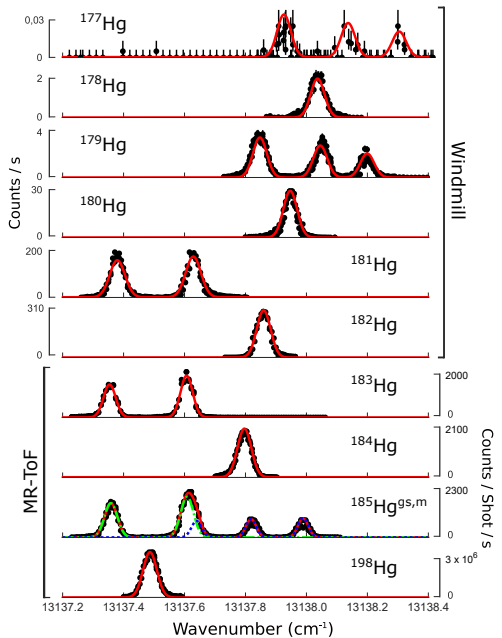


FIG. 7. Examples of the HFS scans taken for the isotopes discussed in this paper. A fit of the data is shown in red. For ^{185}Hg , the green and blue dotted lines correspond to the ground and isomeric state parts of the total fit, shown in red, respectively.

TABLE I. Isotope shifts ($\delta\nu^{198,A}$) and hyperfine splitting parameters (a and b) for the $6s6p^3P_1$ level in mercury atoms and deduced mean-square charge radii ($\delta\langle r^2 \rangle^{198,A}$) and electromagnetic moments (μ and Q) in $^{177-185}\text{Hg}$ isotopes. Results of both optional ground-state spin assignments $I = 7/2$ and $I = 9/2$ for $^{177,179}\text{Hg}$ are shown (see Sec. IV B). The literature data for $\delta\langle r^2 \rangle^{198,A}$ in this table are recalculated from the experimental IS [4].

Isotope	I^π	$\delta\nu^{198,A}$ (MHz)	a (MHz)	b (MHz)	$\delta\langle r^2 \rangle^{198,A} \text{ }^a$ (fm ²)	μ (μ_N)	Q_s (b)	Ref
^{177}Hg	(7/2 ⁻)	54580(390)	-4320(180)	-410(600)	-1.067(8){78}	-1.025(48) ^b	0.57(83)	this work
	(9/2 ⁻)	55170(390)	-3460(180)	-875(600)	-1.083(8){78}	-1.035(60) ^b	1.21(91)	this work
^{178}Hg	0 ⁺	49500(290)	-	-	-0.968(6){71}	-	-	this work
^{179}Hg	(7/2 ⁻)	46310(240)	-3990(80)	-550(200)	-0.905(5){70}	-0.948(24) ^b	0.76(28)	this work
	(9/2 ⁻)	46820(230)	-3150(70)	-1050(210)	-0.915(5){70}	-0.947(27) ^b	1.45(31)	this work
^{180}Hg	0 ⁺	41330(240)	-	-	-0.808(5){60}	-	-	this work
^{181}Hg	1/2 ⁻	5390(280)	15030(120)	-	-0.111(6){11}	0.515(4)	-	this work
		5560(200)	14960(250)	-	-0.114(4){10}	0.5071(7)	-	[2, 4]
^{182}Hg	0 ⁺	33350(260)	-	-	-0.653(5){48}	-	-	this work
^{183}Hg	1/2 ⁻	3100(260)	15190(160)	-	-0.065(5){7}	0.521(6)	-	this work
		3310(100)	15380(130)	-	-0.069(2){6}	0.524(5)	-	[2, 4]
^{184}Hg	0 ⁺	27680(270)	-	-	-0.542(6){40}	-	-	this work
		27720(90)	-	-	-0.544(2){42}	-	-	[4]
^{185}Hg	1/2 ⁻	3350(300)	14930(340)	-	-0.069(6){7}	0.51(1)	-	this work
		3710(30)	14960(70)	-	-0.0764(6){63}	0.509(4)	-	[4]
$^{185}\text{Hg}^m$	13/2 ⁺	27780(190)	-2286(25)	110(300)	-0.543(4){40}	-1.01(1)	-0.15(41)	this work
		27770(110)	-2305(19)	-140(230)	-0.543(2){42}	-1.017(9)	0.20(33)	[4]

^a Statistical errors are given in parenthesis. Systematic errors stemming from the indeterminacy of the F factor (7%) [4] and M_{SMS} are shown in curly brackets (see Eqs. 2-4)

^b Corrected in accordance with hyperfine anomaly estimation (see Sec. III C)

III. Results

A. Extraction of IS and hyperfine splitting parameters

Information on the difference in mean-square charge radius $\delta\langle r^2 \rangle^{A,A'}$ between two nuclei with mass A and A' of the same isotopic chain is extracted from the difference in the positions of the centers of gravity of their respective HFS, ν_0 i.e. their isotope shift of a certain transition. The nuclear electromagnetic moments (magnetic dipole, electric quadrupole) dictate the relative position of the an atomic-state hyperfine-splitting component with respect to ν_0 via the relation

$$\Delta\nu^F = 0.5aK + b \frac{0.75K(K+1) - I(I+1)J(J+1)}{2IJ(2I-1)(2J-1)}, \quad (1)$$

where the dipole and quadrupole hyperfine splitting parameters are given as a and b , $\Delta\nu^F$ represents the energy difference of the hyperfine component with total angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$, with respect to ν_0 [38] and $K = F(F+1) - I(I+1) - J(J+1)$. Fitting of the spectra was performed with the open-source Python package SATLAS [39] and cross-checked with a similar fitting routine in ROOT [37] and the fitting procedure that was used in our previous HFS studies as for instance in [31].

To monitor the stability of the whole system, reference scans of ^{198}Hg were performed regularly. The spec-

tra were fitted separately and the weighed mean of the fit results is taken as a final value. Results of the fits are shown in table I. The experimental errors on the IS include both the fit errors and the spread of individual scan results. For ^{177}Hg , where only a single full spectrum was obtained, the typical dispersion in the extracted HFS centroid position for the other isotopes was added as an additional uncertainty. As the nuclear spin of $^{177,179}\text{Hg}$ could not be determined directly by counting the hyperfine components from the present measurements due to the low angular momentum of the electronic state ($J = 1$) of the upper level of the studied transition (see Fig. 3), we report the IS and hyperfine splitting constant values assuming both possible options for the ground-state nuclear spin of $^{177,179}\text{Hg}$ (7/2 and 9/2, see section IV B). Within the experimental uncertainties, good agreement on the IS, a and b parameters compared to the previous measurements was obtained.

B. Changes in mean-square charge radii

The isotope shift $\delta\nu_i^{A,A'}$ between two isotopes of the same isotopic chain with mass A and A' for transition i , results from the Mass and Field shifts noted $\delta\nu_{M,i}^{A,A'}$ and $\delta\nu_{F,i}^{A,A'}$ respectively:

$$\delta\nu_i^{A,A'} = \nu_i^{A'} - \nu_i^A = \delta\nu_{M,i}^{A,A'} + \delta\nu_{F,i}^{A,A'}. \quad (2)$$

The mass shift can be described as the sum of the so-called normal (NMS) and specific (SMS) mass shifts:

$$\delta\nu_{M,i}^{A,A'} = M \frac{A' - A}{AA'} = (M_{\text{NMS}} + M_{\text{SMS}}) \frac{A' - A}{AA'}, \quad (3)$$

where the NMS is related to the ratio of the electron and proton masses m_e and m_p and to the transition frequency ν_i as $M_{\text{NMS}} = \frac{m_e}{m_p} \nu_i$. The field shift is proportional to an electronic F factor and nuclear parameter $\lambda^{A,A'}$, related to the change in nuclear mean-square charge radius between the two isotopes according to:

$$\delta\nu_{F,i}^{A,A'} = F_{\lambda,i} \lambda^{A,A'} = K(Z) F_{\lambda,i} \delta\langle r^2 \rangle^{A,A'}. \quad (4)$$

In this equation, $\lambda^{A,A'}$ takes into account the influence of the higher-order radial moments. It was shown [40] that the difference between λ and $\delta\langle r^2 \rangle$ is small (less than 10% for heavy atoms) and can be accounted for by the single correction factor $K(Z)$. The F and M factors used, as well as the higher radial moments correction $K(Z)$ were taken from [41], resulting from a combined analysis of data from optical spectroscopy, muonic atoms and elastic electron scattering. The used values are $F_{254 \text{ nm}} = -53(4) \text{ GHz/fm}^2$, $M_{254 \text{ nm}}^{\text{SMS}} = 0 \pm 0.5 M^{\text{NMS}}$ and $K(Z) = 0.927$.

A King plot combines IS for two different atomic transitions to extract information on the electronic F and M factors. The linear relation that exists between the modified isotope shifts $(\delta\nu_i^{A',A} \cdot \frac{AA'}{A'-A})$ following from equations (2)-(4), has a slope κ that equals to the F -factor ratio ($\kappa = F_i/F_{i'}$) of the two transitions i and i' . Information on the M factors can be derived from the intercept with the y-axis, s , via $s = M_{i'} - \kappa M_i$. In [4], this procedure was used to determine F and M factors for the 546-nm transition from the fixed factors for the 254-nm transition. The IS of ^{182}Hg was measured in [4], only for the 546-nm transition. To check the consistency of our data on IS for ^{182}Hg in the 254-nm transition, we include the corresponding point into the King plot from [4] (see Fig. 8). As can be seen in Fig. 8 and its inset, the data from this work matches the previously observed trend.

The isotope shifts obtained from fitting and the resulting calculated differences in mean-square charge radius $\delta\langle r^2 \rangle^{A,A'}$ are shown in table I and plotted in Fig. 9. From $\delta\langle r^2 \rangle^{A,A'}$, the mean-squared deformation parameter $\beta_2 = \langle \beta^2 \rangle^{1/2}$ can be inferred from the relation [38]

$$\langle r^2 \rangle = \langle r^2 \rangle_{\text{DM}}^{\text{spher}} \left(1 + \frac{5}{4\pi} \langle \beta_2^2 \rangle \right), \quad (5)$$

where $\langle r^2 \rangle_{\text{DM}}^{\text{spher}}$ represents the droplet model prediction for a spherical nucleus. The droplet-model calculations have been carried out using the second parametrization of Berdichevsky and Tondeur [42].

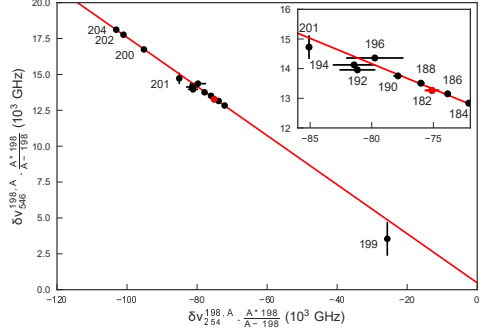


FIG. 8. King plot of the modified isotope shifts in the 546-nm line versus those in the 254-nm line with ^{198}Hg as reference isotope. Inset: zoom around the area of the plot where most of the points are clustered. The new data point for ^{182}Hg fits well to the King-plot line from [4].

C. Electromagnetic moments

The magnetic moments, μ , of the discussed mercury isotopes were calculated using the relation

$$\mu_A = \mu_{A_0} \cdot \frac{I_A}{I_{A_0}} \cdot \frac{a_A}{a_{A_0}} \cdot (1 + {}^{A_0}\Delta^A) \quad (6)$$

where we use $^{199}\text{Hg}^m$ as a reference ($A_0 = 199$, $\mu_{A_0} = -1.0147(8) \mu_N$ [43], $a_{A_0} = -2298.3(2) \text{ MHz}$ [44]). The hyperfine anomaly, ${}^{A_0}\Delta^A$, is defined as

$$A_1\Delta^A = \frac{a_{A_1}}{g_{I,A_1}} \cdot \frac{g_{I,A_2}}{a_{A_2}} - 1 \quad (7)$$

where g_I is the nuclear g factor and the indices A_1 and A_2 refer to two different isotopes with atomic mass numbers A_1 and A_2 . The hyperfine anomaly arises from the differences in charge and magnetization distribution within the nucleus, through the “Breit-Rosenthal” (BR) [45] and “Bohr-Weisskopf” (BW) [46] effects, respectively. If the magnetic hyperfine constant for the point-like nucleus is denoted as a_{point} , then the observed magnetic hyperfine constant a can be presented as follows:

$$a = a_{\text{point}}(1 + \epsilon)(1 + \delta) \quad (8)$$

where ϵ and δ are responsible for the BW and BR effects, respectively. Then, the hyperfine anomaly acquires the simple expression:

$$A_1\Delta^A = A_1\Delta_{\text{BW}}^{A_2} + A_1\Delta_{\text{BR}}^{A_2} = (\epsilon_1 - \epsilon_2) + (\delta_1 - \delta_2). \quad (9)$$

To determine the hyperfine anomaly one should have independent values for magnetic moments and a -constants for the pair of isotopes under study, measured with high

accuracy. In the case of the mercury isotopes, such measurements were done earlier for ten long-lived isotopes and isomers ($^{193-201,193m-199m}\text{Hg}$). Correspondingly, for these nuclei, the hyperfine anomaly is known with sufficient accuracy [47]. Moskowitz and Lombardi [48] have shown that if one neglects the BR part of the anomaly in comparison with its BW component, then the experimental hyperfine anomalies in the 3P_1 atomic state of this series of mercury isotopes, with the odd neutron in the nuclear shell model neutron orbitals $p_{1/2}$, $p_{3/2}$, $f_{5/2}$ and $i_{13/2}$, are well reproduced assuming the simple relation known as the Moskowitz-Lombardi (ML) rule:

$$A_1\Delta_{\text{BW}}^{A_2} = \pm\alpha \left(\frac{1}{\mu_1} - \frac{1}{\mu_2} \right), \quad (10)$$

with sign from $I = \ell \pm \frac{1}{2}$, where $\alpha = 1 \times 10^{-2} \mu_N$ and ℓ is the orbital moment of the unpaired neutron. Neglecting the BR part of the hyperfine anomaly was justified in [49] where $A_1\Delta_{\text{BR}}^{A_2}$ was calculated with a diffuse nuclear charge distribution. In particular, according to [49], $^{199}\Delta_{\text{BR}}^{201} = -1.8 \times 10^{-4}$ whereas $^{199}\Delta_{\text{BR}}^{201} = -1.5 \times 10^{-3}$ [47]. The ML-rule was further supported by calculations from a microscopic theory [50]. We applied the ML-rule to estimate the BW-correction for the magnetic moments of $^{177,179}\text{Hg}$, taking into account the description of the experimental hyperfine anomaly by this rule for the variety of neutron single-particle states in mercury nuclei with the mass spanning a rather large range. For previously-measured isotopes and isomers the maximal deviation of the experimental $^{199}\Delta_{\text{BW}}^A$ from the ML-calculation is equal to 2.5×10^{-3} . We conservatively estimated the error of ML-prediction for the hyperfine anomaly in $^{177,179}\text{Hg}$ as 5×10^{-3} . It was shown in [51] that $A_1\Delta_{\text{BR}}^{A_2}$ is proportional to $\delta\langle r^2 \rangle^{A_1, A_2}$. Thus, $^{199}\Delta_{\text{BR}}^A$ for $^{177,179}\text{Hg}$ can be estimated by scaling the calculated $^{199}\Delta_{\text{BR}}^{201}$ [49]. The uncertainty of this correction was estimated to be 10%. For $^{177,179}\text{Hg}$, the BR correction are $^{199}\Delta_{\text{BR}}^{177} = -1.7 \times 10^{-3}$, $^{199}\Delta_{\text{BR}}^{179} = -1.5 \times 10^{-3}$. Assuming an $I = 7/2$ assignment, $^{199}\Delta_{\text{BW}}^{177} = -1.2 \times 10^{-4}$ and $^{199}\Delta_{\text{BW}}^{179} = 7 \times 10^{-4}$. Under the $I = 9/2$ assignment, $^{199}\Delta_{\text{BW}}^{177} = -1.9 \times 10^{-2}$ and $^{199}\Delta_{\text{BW}}^{179} = -2.0 \times 10^{-2}$. These corrections as well as the increase of uncertainties according to the aforementioned prescriptions, are taken into account in Table I.

The spectroscopic quadrupole moments were calculated using the relation

$$Q_s^A = Q_s^{A_0} \frac{b_A}{b_{A_0}} \quad (11)$$

with the reference values for ^{201}Hg : $Q_s^{A_0} = 0.387(6)$ b, $b_{A_0} = -280.107(5)$ MHz taken from [52] and [53], respectively. The resulting spectroscopic quadrupole moments are shown in table I.

IV. Discussion

A. Changes in mean-square charge radii, shape staggering and ‘return to sphericity’ of light mercury isotopes

The change in the nuclear mean-square charge radius, $\delta\langle r^2 \rangle$, for lead [54–58] and mercury ([4] and this work) isotopes are plotted with respect to $N = 126$ at the top of Fig. 9. Three distinctly different regions are observed in the mercury charge radii. Mercury isotopes with $N > 105$ follow a smooth trend, identical to the one of the isotopic chain of lead. At $100 < N < 106$, in the neutron mid-shell region between the closed shells at $N = 82$ and $N = 126$, a large shape staggering is observed. Here, ground-state radii of the odd- A mercury isotopes deviate substantially from the trend of the lead isotopes, which were found to keep their near-spherical shape at and beyond the neutron midshell [56, 57]. See also Fig. 9 b), where the difference in $\delta\langle r^2 \rangle$ between mercury and lead isotones is shown. From the data obtained in the present work, it is observed that the staggering stops at ^{180}Hg and $\delta\langle r^2 \rangle$ for mercury isotopes returns to the trend of lead. In-beam recoil-decay tagging measurements, [10–12, 59–61], showed that the band-head energies of strongly-prolate deformed intruder bands in even- A isotopes increase rapidly for mercury isotopes with decreasing neutron number at $N < 101$. The in-beam studies by Kondev *et al.* [60] and ^{181}Pb α -decay analysis by Jenkins *et al.* [16] have shown that a pronounced structural change takes place when moving from $^{181,183}\text{Hg}$ to ^{179}Hg . Based on their decay scheme deduced in [60], the authors proposed that the ground state of ^{179}Hg is near-spherical with a possible weak-prolate deformation, rather than a strong, prolate deformation. A similar interpretation was proposed for lighter odd- A mercury isotopes with $A = 173 - 177$ [12, 13, 62]. While those different studies had shown some indications of the shape of the ground state, our data provide a direct measurement of the ground-state charge-radii changes. The return of the lightest mercury nuclei to the trend of the weakly deformed mercury isotopes with $N > 105$ (and to the near-spherical trend in lead nuclei) delineates the region of shape staggering to near the neutron mid-shell region at $100 < N < 106$.

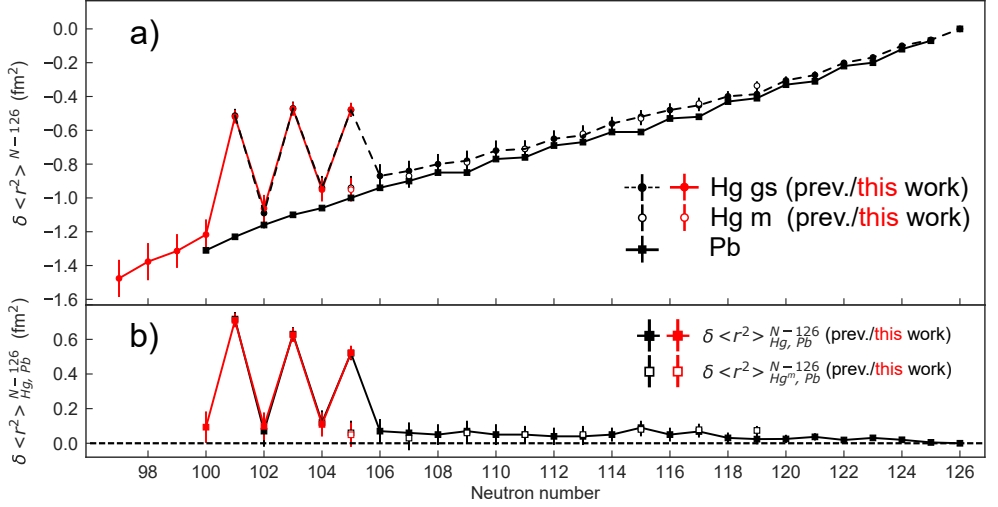


FIG. 9. a) Comparison of the change in mean-square charge radius for mercury and lead isotopic chains as a function of neutron number N , using $N = 126$ as a reference.

b) Difference between the mercury and lead changes in charge radii where $\delta \langle r^2 \rangle_{\text{Hg}, \text{Pb}}^{N-126}$ is defined as $\delta \langle r^2 \rangle_{\text{Hg}}^{N-126} - \delta \langle r^2 \rangle_{\text{Pb}}^{N-126}$. Data points shown in red and black correspond to this work and previous work respectively ([58] for lead, [4] for mercury).

B. Magnetic moments and spins of $^{177,179}\text{Hg}$

The ground-state spin and parity of ^{179}Hg was previously assigned $I^\pi = (7/2^-)$, based on experimental data obtained for the α -decay of ^{183}Pb and subsequent α -decays to daughter (^{175}Pt) and grand-daughter (^{171}Os) nuclei [11, 16]. The same assignment, $I^\pi = (7/2^-)$, was proposed for the ground state of ^{177}Hg , based on decay properties of the $13/2^+$ isomeric state in this nucleus [12] and α -decay of ^{181}Pb [20]. As was indicated in Sec. III A, we also tested $I = 9/2$ as a possible assignment, since the $\nu f_{7/2}$ and $\nu h_{9/2}$ orbitals are assumed to play a dominant role in the negative-parity states around $N = 97, 99$. The quality of fitting is the same for both assumptions. However, the measured large and negative magnetic moments of $^{177,179}\text{Hg}$ rule out a $\nu h_{9/2}$ hole configuration as this is expected to give a large positive magnetic moment $\mu_{\text{th}}(\nu h_{9/2}) = +0.69 \mu_N$ [63].

Two plausible origins of the $^{177,179}\text{Hg}$ spins and magnetic moments are considered:

- They arise from the strong Coriolis mixing of the Nilsson states of $\nu f_{7/2}$ and $\nu h_{9/2}$ parentage at very low deformation ($\beta_2 \approx 0.05 - 0.07$) or
- they may be regarded as spherical $\nu f_{7/2}$ -hole states.

Let us first focus on the explanation a).

In [11], two likely candidates for the ground-state Nilsson configuration of ^{179}Hg were proposed: the $7/2^-$ [514] and $7/2^-$ [503] orbitals, arising from the $\nu h_{9/2}$ and $\nu f_{7/2}$

neutron shell-model states respectively. These orbitals come close to the Fermi level only for small prolate deformations ($\beta_2 < 0.15$). In ^{175}Pt , the α -decay daughter of ^{179}Hg , the $\nu h_{9/2}$ $7/2^-$ [514] orbital was chosen as preferable for the ground state band [12]. However, several measured magnetic moments of the $7/2^-$ [514] Nilsson state of deformed $N = 105$ nuclei ($^{183}\text{Pt}^m$, ^{177}Hf , ^{175}Yb) are close to $\mu \approx +0.8 \mu_N$ [64]. Nilsson-model calculations describe these experimental data fairly well [65]. The calculations with the same approach predict positive moments for $^{177,179}\text{Hg}$ even at rather low deformation ($\mu(7/2^-$ [514], $^{179}\text{Hg})_{\text{th}} = +0.46 \mu_N$ and $+0.34 \mu_N$ at $\beta_2 = 0.15$ and 0.10 respectively). Coriolis-mixing might however play a role as the lowest states in the lightest mercury isotopes display a high spin at low deformation. At $\beta_2 < 0.15$, contributions of the different Nilsson-orbitals stemming from the $\nu f_{7/2}$ and $\nu h_{9/2}$ orbitals to the lowest $7/2^-$ state become nearly equal. This mixing would bring the magnetic moment down in value with respect to a pure $7/2^-$ [514] configuration and might be a possible explanation of the measured magnetic moments of $^{177,179}\text{Hg}$.

Let us now discuss option b) and explore the interpretation of the ground-states of $^{177,179}\text{Hg}$ as spherical $\nu f_{7/2}$ -hole states.

Magnetic moments of $^{177,179}\text{Hg}$ come comparatively close to the single particle estimation for spherical neutron shell $\nu f_{7/2}$: $\mu_{\text{s.p.}} = -1.3 \mu_N$ (with the com-

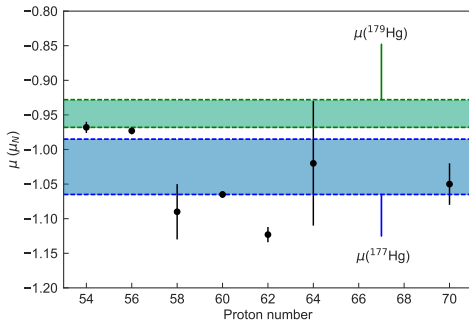


FIG. 10. Comparison of magnetic moments for $N = 83$ isotones, having one neutron in the $(\nu f_{7/2})$ shell above the closed shell $N = 82$, with the measured $\mu(^{177}\text{Hg})$ and $\mu(^{179}\text{Hg})$ indicated by shaded boxes. Data from [64], [66] and references therein.

monly adopted renormalization of the neutron g factors: $g_s^{\text{eff}} = 0.6g_s^{\text{free}}$ and $g_l^{\text{eff}} = -0.05$). It is instructive to compare the magnetic moments of the presumed $7/2^-$ $^{177,179}\text{Hg}$ ground states with measured magnetic moments of the ground states of $N = 83$ isotones with one neutron in the $f_{7/2}$ shell. This comparison is shown in Fig. 10. One can see that $\mu(^{177,179}\text{Hg})$ corresponds to $\mu(\nu f_{7/2})$ in the $N = 83$ isotones for which all show a rather large negative magnetic moment value. If this interpretation is valid, then the ground states of $^{177,179}\text{Hg}$ could be regarded as holes in the $\nu f_{7/2}$ orbital within a simple shell model picture.

This means that for the light mercury isotopes, the state arising from a neutron hole in the $\nu f_{7/2}$ orbital lies above that arising from a neutron hole in the $\nu h_{9/2}$ orbital. As a consequence, the state ordering for $Z = 80$ and $N < 100$ is reversed with respect to the $N = 83$ isotones in the vicinity of the stable isotopes, where the $\nu f_{7/2}$ orbital is filled first after the $N = 82$ shell closure. Surprisingly, with the increase of Z after ^{80}Hg , the normal ordering is restored in ^{81}Tl and ^{82}Pb . The ground-state spin and parity of $^{181}\text{Pb}_{99}$ was determined as $9/2^-$ due to a hole in $\nu h_{9/2}$ shell arising from the complete depletion of the $i_{13/2}$ and $p_{3/2}$ shells lying above the $N = 100$ spherical subshell closure [20]. Similarly, the odd neutron state in $^{180}\text{Tl}_{99}$ was assumed to be a $\nu h_{9/2}$ -hole state on the basis of its magnetic moment value [17, 67]. Thus, for $Z > 80$ the $\nu h_{9/2}$ shell appears to be situated above the $\nu f_{7/2}$ shell.

It was shown that the energy differences between the lowest-lying $9/2$ and $7/2$ states for the $N = 83$ and $N = 85$ isotones show a rapid drop above $Z = 64$ (see [68] and references therein). According to Bianco *et al.* [68], this drop reflects the gradual approach in energy of the $\nu h_{9/2}$ and $\nu f_{7/2}$ neutron single-particle orbitals. The presumed convergence of the $\nu h_{9/2}$ and $\nu f_{7/2}$ neutron levels

also provides a natural explanation for the anomalous absence of charged-particle emission from the high-spin isomer of ^{160}Re [69]. This shell evolution was explained in [68] by the influence of the tensor part of the nucleon-nucleon interaction [70]. It was predicted in Ref. [68] that the energies of the neutron single-particle orbitals may become inverted for high Z . The interpretation of the $^{177,179}\text{Hg}$ ground states as $\nu f_{7/2}$ shell-model states and their first excited states as predominantly $\nu h_{9/2}$ states [11, 12] is in agreement with this description.

C. Quadrupole moments of $^{177,179}\text{Hg}$

The quadrupole moments of $^{177,179}\text{Hg}$ extracted from their respective HFS b constants are consistent with a simple spherical shell model approach. According to the seniority scheme [71], quadrupole moments should be linearly dependent on the number of particles occupying a certain orbital as can be seen from equation 12.

$$\langle j^n | \hat{Q} | j^n \rangle = \frac{2j+1-2n}{2j+1-2\nu} Q_{s.p.} \quad (12)$$

$$Q_{s.p.} = -e \frac{2j-1}{2j+2} \langle r^2 \rangle_j \quad (13)$$

Here, a j^n configuration with n nucleons is labelled by a seniority ν , the number of unpaired neutrons ($\nu = 1$ in our case). The observed spectroscopic quadrupole moment Q_s is represented by $\langle j^n | \hat{Q} | j^n \rangle$, while $Q_{s.p.}$ indicates the single-particle quadrupole moment value. Recently, the seniority scheme with a linear dependence of Q_s on the number of particles in the filling of a shell has been found to work remarkably well for the cadmium, astatine and actinium isotopes [36, 72, 73]. One can check the validity of the seniority scheme for the filling the $\nu f_{7/2}$ orbital with the assumption that $^{177,179}\text{Hg}$ have 5 and 7 neutrons in this shell ($n = 5, 7$), respectively. For $n = 1, 3, 5$ we choose $^{145,147,149}\text{Sm}$ [64]. The values of $Q(^{177,179}\text{Hg})$ were scaled according to Eq. 13 by 16% using the global evaluation of $\langle r^2 \rangle$ data in [58], to remove the dependence of Q on $\langle r^2 \rangle$, which distorts the presumed linear dependency.

As can be seen from Fig. 11, the light mercury isotopes follow the trend given by samarium even though the considered nuclei have very different neutron numbers: $N = 83-87$ for samarium and $N = 97-99$ for mercury. This observed correspondence of the $Q_s(^{177,179}\text{Hg})$ values to the seniority-scheme prediction supports the assumption of the $\nu f_{7/2}$ -hole nature of the $^{177,179}\text{Hg}$ ground states.

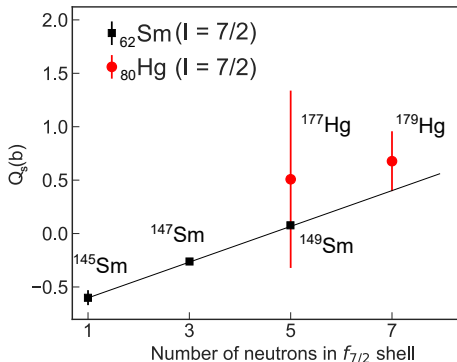


FIG. 11. Comparison of quadrupole moments for the $^{145,147,149}\text{Sm}$ isotopes (data from [64]), with measured values for $^{177,179}\text{Hg}$ corrected for the difference in charge radius between samarium and mercury.

D. Comparison with Nuclear Density Functional Theory (DFT) and Monte-Carlo Shell Model (MCSM) calculations

Frauendorf and Pashkevich [6] described shape staggering in the mercury charge radii using a microscopic-macroscopic approach with Strutinsky shell corrections, thereby proving that mean-field models are able to predict the shape staggering and shape coexistence in mercury. More recently, the even-even mercury isotopes were studied and spectroscopic observables (e.g. charge-radii) were calculated using an Interacting Boson Model with configuration mixing (IBM-CM) [74] and a beyond mean field approach [75]. In both approaches, the deformation energy surfaces for nuclei near the midshell have a lowest-energy minimum at prolate deformation, accompanied by a second, oblate minimum (for $176 < A < 186$ in [75] and $180 < A < 186$ in [74]). This is in contrast to experimental results, that indicated that the even-even mercury isotopes have weakly deformed (oblate) ground-state shapes [7]. Both papers [74, 75] only discuss the even- A mercury cases and do not go deeper into the origin of the experimentally observed shape staggering in the odd- A mercury isotopes or the prediction of its magnitude.

Early DFT calculations for both even- and odd-mass isotopes mercury isotopes with the SLy4 parameterization achieved a reproduction of the odd-even radius staggering for $N > 100$, by fitting the pairing strength to the one-particle separation energy of these isotopes [76]. In [77] the shape coexistence in the region was confirmed as a mechanism for the observed radius staggering for the SLy4, Sk3 and SGII parameterizations with fixed gap parameters, although the exact staggering could not be reproduced. In [78], the SLy4 parameterization was

again employed, but this time with a pairing strength adjusted to the odd-even staggering of the lead isotopes, leading to a qualitative correct staggering that is off by four neutron numbers. As explicitly noted in [78], the precise staggering pattern depends sensitively on the details of the effective interaction. This can be explicitly demonstrated by comparing results for the SLy4 parameterization from all three sources, [76, 77] and [78], which differ in the pairing interaction employed in the respective calculations. Altogether, the past DFT investigations led to an overall understanding of the physics of the phenomenon. However, because of the known deficiencies of the available parameterisations of the functional, achieving a quantitative description of all its details is still not possible.

In an attempt to understand the behavior exhibited by the light mercury isotopes, and to extend the description to odd- A mercury isotopes, Density Functional Theory (DFT) and large-scale Monte-Carlo Shell model (MCSM) calculations were performed in this work which are described in sections IV D 1 and IV D 2 respectively.

1. DFT calculations

In a mean-field picture, a dramatic staggering of the isotope shift as observed in experiment can be achieved as a consequence of a ground-state *shape* staggering. In order for a shape staggering to occur and produce a large change in nuclear charge radius, three conditions need to be met. First, the isotopes must have multiple competing shapes, which in the case of neutron-deficient mercury isotopes, are oblate, weakly prolate and strongly prolate. Second, the minima must exhibit sufficiently different deformations, leading to substantial differences in the corresponding calculated root-mean square radius. For the mercury isotopes, the even- A isotopes should have weakly-deformed minima, while the odd- A nuclei in the region should have strongly-prolate deformations. Third, the excitation energy of the strongly-prolate minimum should be small for a specific set of nucleon numbers, $101 \leq N \leq 105$. More precisely, it should be comparable to the odd-even *mass* staggering for these isotopes i.e the difference in binding energy between odd- A isotopes and their even- A neighbours.

In this way, the difference in odd-even mass staggering in both wells can shift the energetic balance between the weakly- and strongly-deformed configurations. As is argued below, these conditions put extremely stringent constraints on the parameters of the DFT functionals, far beyond the precision with which these parameters have been determined so far.

Two sets of systematic calculations for the mercury isotopes were performed. The first set considers axially-symmetric configurations obtained using the HFBTHO code [79, 80], for six different Skyrme functionals UNEDF0 [81], UNEDF1 [82], UNEDF1^{SO} [83], SLy4 [84],

SkM* [85] and SGII [86].

The second set is comprised of 3D coordinate-space calculations with the MOCCA code [87], employing the eight parameterizations of the recent SLy5sX [88] family supplemented by a zero-range surface pairing interaction [89] with a standard pairing strength [90]. In both sets of calculations, we retained the blocked configurations for the odd- A isotopes that are lowest in energy, which in general do not correspond to the experimental ground state spins and parities.

None of these fourteen parameterizations predict potential energy surfaces (PES) that fulfill all three conditions referred to above. However, all of them at least predict near-degenerate oblate and prolate minima to co-exist in light mercury isotopes. This feature thus seems to be a generic property of the bulk macroscopic energy, and is independent of the detailed orderings of single-particle levels that differ between functional parameterizations. In particular, the orbitals corresponding to the experimental ground-state spins and parities do not appear at the Fermi surfaces of odd- A mercury isotopes.

We will focus in what follows on four different parameterizations. The first, UNEDF1^{SO}, is based on the UNEDF1 parameterization which has been adjusted to global nuclear properties across the nuclear chart, but with modified spin-orbit and pairing properties to reproduce detailed spectroscopic properties of nuclei around ²⁵⁴No. Calculations here have been carried out for three different variants: one employing the pairing prescription from [83], and two others with an amplified pairing strength compared to the original parameterization, by 8% and 20% respectively.

The three remaining functionals, SLy5s1, SLy5s4 and SLy5s8, are members of a family of parameterizations constructed on the basis of SLy5* [91] to study the impact of varying the surface tension of Skyrme functionals. SLy5s8 has a surface tension similar to SLy4 and SLy5*, while for SLy5s7 down to SLy5s1, it takes progressively smaller values. Because of the nature of the fitting protocol, all members of the family exhibit similar single-particle structures. However, the deformation properties of the parameterizations are quite different, which is a consequence of the differences in surface tension. In particular, the parameterization with lowest surface tension, SLy5s1, gives quite a satisfying description of the fission barriers of heavy nuclei, such as actinides and ¹⁸⁰Hg [92]. Here, results are presented for SLy5s1, SLy5s4 and SLy5s8 representing functionals with low, intermediate and high surface tension respectively.

The PES for the even- A neutron-deficient mercury isotopes as calculated with UNEDF1^{SO} with standard pairing as a function of the axial quadrupole deformation, are shown in Fig. 12. For all isotopes, the lowest minimum is the oblate one. In a narrow region, $86 \leq N \leq 106$, the strongly-prolate minimum is very close in energy to both the strongly-prolate and the oblate minimum. In this way, the UNEDF1^{SO} PES fulfills the first two conditions for producing a sharp radius staggering. The other pa-

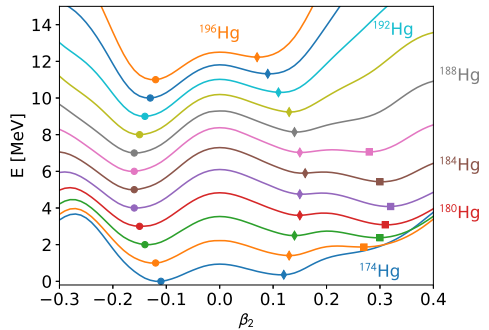


FIG. 12. Potential energy surfaces calculated using the UNEDF1^{SO} functional for even- A mercury isotopes between $A = 174$ and $A = 196$ as a function of the axial quadrupole moment, β_2 . Energies are renormalized to the minimum of the curve, with an additional offset of 1 MeV between isotopes. Circles indicate the oblate minima, diamonds the weakly prolate minima, and squares the strongly-prolate minima (if present).

rameterizations discussed here give qualitatively similar energy curves with the same overall pattern of minima, but with slightly different relative energy between them. Albeit small, these differences can change the energetic order of minima when going from one parameterization to the next.

To appreciate the detailed energy balance between the different configurations in both odd- and even- A isotopes, the excitation energies of the weakly- and strongly-prolate minima are shown in Fig. 13 with respect to the oblate minimum for UNEDF1^{SO} with different values of the pairing strength, as well as for SLy5s1, SLy5s4 and SLy5s8. An unaltered UNEDF1^{SO}, Fig. 13, predicts all isotopes between $99 \leq N \leq 103$ to be strongly-prolate deformed. Increasing the pairing strength by 8% suffices to change the predicted deformation of the even- A isotopes in that region, producing a staggering pattern. Further increasing the pairing strength makes the shape staggering completely disappear: the oblate minimum is the lowest for all isotopes. For SLy5s1 as well, the ground state staggers between strongly-prolate and oblate minima in the region $99 \leq N \leq 103$. By increasing the surface tension the shape staggering can be changed: for SLy5s4 only two odd isotopes exhibit strong prolate deformation while none do for SLy5s8.

For the SLy5sX parameterizations, larger surface tension penalizes the strongly-prolate minimum compared to the oblate and weakly-prolate ones because of its larger deformation. Increasing the pairing strength serves a similar purpose: the strongly-prolate minimum gains in energy compared to the oblate minimum. Note that for all variants of UNEDF1^{SO} the size of the odd-even mass staggering in the strongly-prolate minimum is almost in-

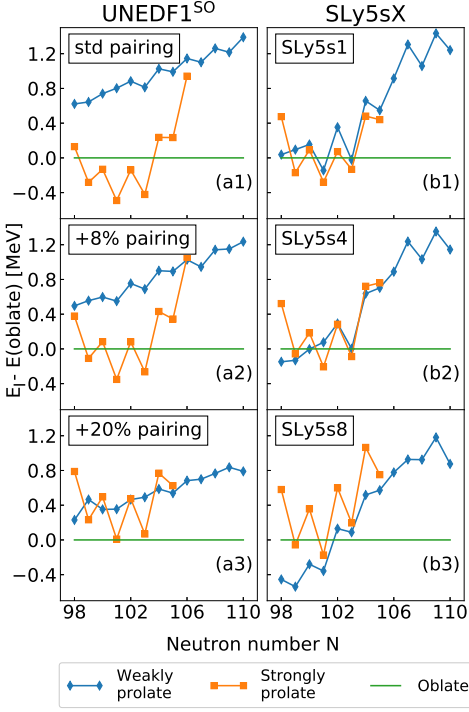


FIG. 13. Differences between energies of weakly and strongly prolate configurations and those corresponding to the oblate configurations, as calculated with UNEDF1^{SO} (left) and the SLy5sX parameterizations (right). For UNEDF1^{SO} calculations with standard pairing (top), pairing amplified by 8% (middle) and pairing amplified by 20% (bottom) are presented. For the SLy5sX family, results for SLy5s1 (top), SLy5s4 (middle) and SLy5s8 (bottom) are presented.

dependent of the size of the pairing strength: the mass staggering is at the root of the shape staggering, but the detailed fine tuning of the staggering is achieved by changing the balance of the minima and not through a change of the size of the odd-even mass staggering. Although being quite different features of the effective interaction, the variation of the pairing strength and the surface tension can both be employed to change the balance of minima in order to fine-tune the shape staggering.

The effect of the shape staggering on the calculated radius staggering is shown in Fig. 14 for the set of UNEDF1^{SO} calculations and in Fig. 15 for the SLy5sX calculations. Large charge radii appear where the calculated ground state has a strong, prolate deformation. SLy5s1 and UNEDF1^{SO} with 8% increase in pairing strength predict a staggering pattern that sets in later

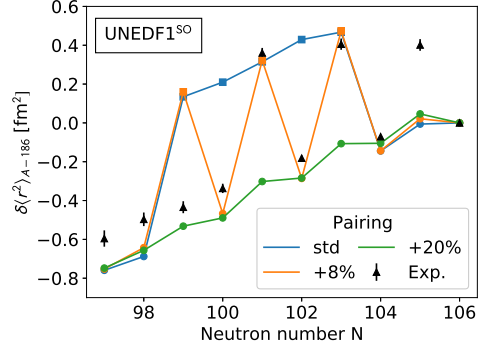


FIG. 14. Comparison of the calculated isotopic shift using the UNEDF1^{SO} parameterization for three different values of the pairing strength with experimental data, normalized to ¹⁸⁶Hg. As in Fig. 12 and Fig. 13, circles indicate oblate, diamonds weakly-prolate and squares strongly-prolate configurations.

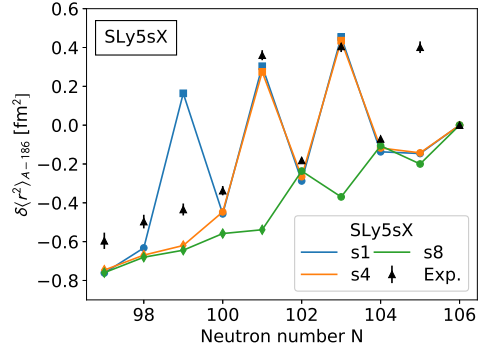


FIG. 15. Same as Fig. 14, but for the SLy5s1, SLy5s4 and SLy5s8 parameterizations. Circles, diamonds and squares indicate oblate, weakly-prolate and strongly-prolate configurations, respectively.

with reducing neutron number.

The presented DFT results show that the origin of the radius staggering can be properly identified in terms of the ground-state shape staggering between the oblate (or strongly-prolate) and strongly-prolate configurations along the chain of mercury isotopes. The effect is extremely sensitive to the fine details of the functional: by slightly varying the pairing strength of (UNEDF1^{SO}) or the surface tension (SLy5sX) of the functional, one can fine tune the detailed balance between the three, coexisting minima, which can change dramatically the shape staggering and hence the radius staggering.

While this effect is demonstrated here for the pair-

ing strength and surface tension, the balance between the minima also depends on many other properties of the functionals both in the particle-hole and particle-particle channels. Because of this sensitivity, it is very unlikely that any of the existing parameterizations can be tuned to reproduce exactly this very particular shape-coexistence pattern. For this reason, we can not meaningfully state a strong preference for any parameterization discussed here. The precise staggering pattern could, however, be used to impose stringent conditions on future parameter fits.

2. MCSM calculations

Monte Carlo Shell Model (MCSM) calculations are a type of configuration-interaction approach for atomic nuclei that uses the advantages of quantum Monte-Carlo, variational and matrix-diagonalization methods [93]. This is the first time such calculations were performed for such a heavy system as the mercury isotopes and they are the heaviest MCSM calculations so far. The massively parallel K-supercomputer [94] provided the computing power to execute the calculations.

The model-space single-particle orbitals used in these calculations consist of proton orbitals from $1g_{7/2}$ up to $1i_{13/2}$ and neutron orbitals from $1h_{9/2}$ up to $1j_{15/2}$, using the doubly-magic ^{132}Sn nucleus as inert core. As a result, a large number of nucleons (30 protons and up to 24 neutrons) were left to interact in a large model space.

In these orbitals, all nucleons interact through effective nucleon-nucleon (NN) interactions. The neutron-neutron ($n-n$) and proton-proton ($p-p$) interactions are taken from [95], while the proton-neutron ($p-n$) interaction from [96] was used. Effective charges for protons and neutrons [97] being $1.6e$ and $0.6e$ were used together with a spin-quenching factor of 0.9 [97] and single-particle energies were adjusted to properties of doubly-magic ^{132}Sn and ^{208}Pb nuclei.

Eigenstates were calculated for mercury isotopes with $177 \leq A \leq 186$ with excitation energies below 2 MeV and spins and parities as observed in experiment. For each of these eigenstates, the magnetic moment, quadrupole moment, excitation energy and nucleon occupation numbers were computed.

As shown in Fig. 16, the MCSM calculations reproduce the magnetic and quadrupole moments for both experimentally-observed ground states and isomeric states of all measured odd- A isotopes. While for some isotopes the electromagnetic moment data clearly favor one eigenstate over the others, such a distinction cannot be made for all isotopes. The magnetic moment of ^{177}Hg for instance clearly favors the $7/2_1^-$ state over $7/2_2^-$, while in ^{181}Hg , the magnetic moment of all $1/2^-$ states is nearly identical. MCSM calculations were also performed for $9/2^-$ states in $^{177,179}\text{Hg}$. The magnetic moments of these states were found to be large and positive: $\mu(9/2^-) = 1.26, 0.72$ and $1.26, 1.47 \mu_N$ for the

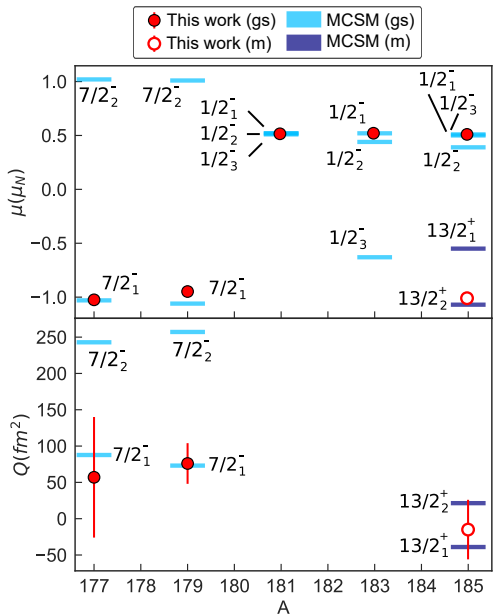


FIG. 16. Comparison of experimental magnetic moment values from laser spectroscopy with MCSM calculations for odd- A neutron-deficient mercury isotopes ($A = 177 - 185$). Red circles refer to experimental data.

first two excited states with $I^\pi = 9/2^-$ in $^{177,179}\text{Hg}$, respectively.

The MCSM eigenstate is given by a superposition of MCSM basis vectors. Each MCSM basis vector is a deformed Slater determinant, for which intrinsic quadrupole momenta Q_0 and Q_2 can be calculated. Q_0 and Q_2 can be used as partial coordinates, from which the nuclear shape parameters β_2 and γ can also be extracted by standard relations [98]. A given MCSM basis vector is placed as a circle on the Potential Energy Surface (PES) according to Q_0 and Q_2 . The importance of this basis vector in the eigenstate is expressed by the area of a circle, being proportional to the square of the overlap to the eigenstate. This is called the T-plot [99].

The calculated β_2 values are extracted via the following expression:

$$Q_0 = \frac{3Z}{\sqrt{5}\pi} R^2 \langle \beta_2 \rangle (1 + 0.36 \langle \beta_2 \rangle) \quad (14)$$

where R^2 is calculated as $R^2 = (1.2 \text{ fm } A^{1/3})^2$ [38]. Since the mass quadrupole moment, rather than the electric quadrupole moment results from MCSM calculations, the factor Z was replaced in Eq. 14 by A . The correction

term $0.36\langle\beta_2\rangle$ was neglected, because $\langle\beta_2\rangle$ is at most about 0.2 presently. The resulting equation becomes

$$\langle\beta_2\rangle = Q_0 \left(\frac{\sqrt{5}\pi}{3AR^2} \right) \quad (15)$$

corresponding to the equations used in [100, 101].

The differences in mean-square charge radii $\delta\langle r^2 \rangle$, normalized to ^{186}Hg , from experimentally measured isotope shifts were compared to MCSM calculations using the extracted β_2 values (Eq. 15) in combination with the nuclear droplet model [42] by Eq. 5 as detailed in [38, 102]. Panel a) of Fig. 17 shows the comparison of MCSM calculations with experimental values.

The ground state of ^{186}Hg is known from its observed level scheme and from Coulomb-excitation measurements to be only weakly deformed [7, 103]. This nucleus was used to normalize the experimental $\delta\langle r^2 \rangle_{A-186}$ -values from laser-spectroscopy measurements with theoretical calculations. The plot in panel a) of Fig. 17 highlights $\delta\langle r^2 \rangle_{A-186}$ of the lowest-lying nuclear states with the correct spin and parity in a light blue shade if the

calculated magnetic moment is similar to the experimentally observed one and in gray when they do not match. Since even-even nuclei, having 0^+ ground states, do not have a magnetic moment, all even-mass states are indicated in blue on Fig. 17. The width of the colored areas corresponds to the spread of MCSM basis-vector deformation parameters on the PES. The levels for which both the magnetic moment and deformation agree with experiment are connected by the blue band.

An overall agreement for the shape staggering is observed in both the magnitude and location as a function of neutron number. In all but $^{181,185m}\text{Hg}$ the state corresponding to the electromagnetic moments and charge-radii differences observed in experiment is also the lowest state of that given spin and parity. The difference in energetic ordering for $^{181,185m}\text{Hg}$ might come from the limit of 24 MCSM basis vectors used in the calculation. Dedicated calculations with 12 and 16 basis vectors showed indeed that the magnetic and quadrupole moments converged to a large extent for 24 basis vectors, while the excitation energy of the calculated states could still shift when more basis vectors would be added.

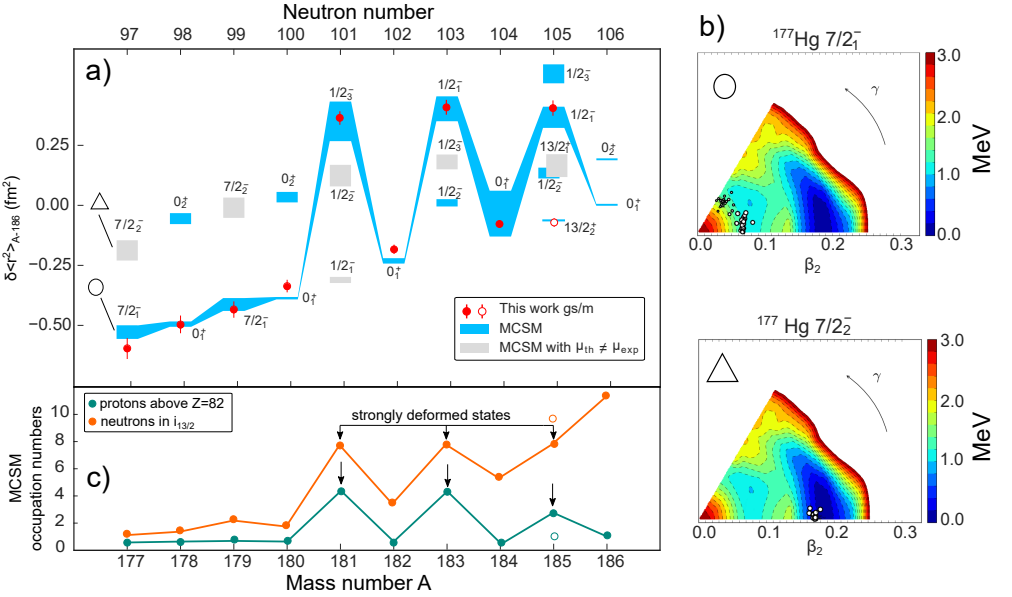


FIG. 17. a) : Mean square charge radius relative to that of the ground state of ^{186}Hg . Red points are experimental data from this experiment. The blue shaded area indicates the radii corresponding to MCSM eigenstates for the observed ground states, $J^\pi = 0^+$ for even- A , $1/2^-$ for $^{181,183,185}\text{Hg}$, and $7/2^-$ for $^{177,179}\text{Hg}$. In addition, the $\delta\langle r^2 \rangle$ value of the isomeric state $13/2_2^+$ is shown by the lower line for ^{185}Hg . The width of the blue shaded areas corresponds to the spread of MCSM basis-vector deformation parameters on the PES. The gray areas show MCSM eigenstates for which the calculated magnetic moments differ from measured values. b): T-plots for two states in ^{177}Hg are shown on the right (see text for more information). c): Proton and neutron occupation numbers.

For the lightest mercury isotopes with $A \leq 180$, the MCSM correctly describes the trend towards sphericity, magnetic moments and even the energetic ordering of levels. The unusually rapid and large change of the $\delta < r^2 >_{A=186}$ value for neutron-deficient mercury isotopes with $A < 186$ is thus reproduced well in current calculations. While the present Hamiltonian is rather standard, the unprecedented size of the configuration space plays an essential role in the calculation outcome.

Shape staggering mechanism

The present MCSM calculation enables one to investigate the underlying microscopic factors driving the abrupt change between the near-spherical and deformed states. Notably, the change in nuclear shape is related to a change in the occupation numbers. The most important ones are the proton $1h_{9/2}$ orbital, which is the strongest contributor to protons excitation above $Z = 82$, and the neutron $1i_{13/2}$ orbital, shown in panel c) of Fig. 17. Large and constant values (≈ 8) of the occupation number of the neutron $1i_{13/2}$ orbital are observed for the strongly deformed $1/2^-$ states of $^{181,183,185}\text{Hg}$. In addition, more than 2 protons are excited above the $Z = 82$ closed shell to the $1h_{9/2}$ orbital. For weakly-deformed states, the occupation number of the $1i_{13/2}$ orbital grows steadily with neutron number, while that of the proton $1h_{9/2}$ orbital remains small as is expected from the usual filling of proton and neutron orbitals. The origin of this abrupt change in occupancy numbers as a function of neutron number is found in the monopole component of the NN interaction. The effect of the monopole interaction between protons in the orbital j_p and neutrons in the orbital j_n can be expressed as

$$E_{\text{mon}} = f(j_p, j_n) n_\pi(j_p) n_\nu(j_n), \quad (16)$$

where $f(j_p, j_n)$ is the monopole matrix element, $n_\pi(j_p)$ and $(n_\nu(j_n))$ stand for the number of protons and neutrons in the specified orbitals, respectively. The average value of $f(j_p, j_n)$ for different orbitals is about -0.2 MeV in mercury, but $f(\pi h_{9/2}, \nu i_{13/2})$ stands out with a strongly-attractive value of -0.35 MeV. This is due to the similarity in radial wave functions of the two orbitals and due to the effect of the tensor force originating from the attractive $j_>-j_<$ coupling [104]. Once optimal numbers of protons and neutrons are found in

these orbitals, they produce a large quadrupole deformation, resulting in an increased binding energy due to the proton-neutron quadrupole correlation energy. However, these orbitals lie above the Fermi energy and a mechanism is needed to bring such states down in energy. The strong attractive nature of the monopole interaction provides such a mechanism for $f(\pi h_{9/2}, \nu i_{13/2})$ since it is more attractive by 0.15 MeV than the average interaction. When moving from ^{180}Hg to ^{181}Hg , three more protons and six more neutrons in those orbitals, produce an additional monopole binding energy of 2.7 MeV besides the quadrupole contribution. The monopole interaction thus shifts the strongly-deformed state down in energy. The influence this monopole interaction has on the PES is shown in Fig. 18 where the strength of this interaction is artificially reduced. Here, a shift of the minimum, indicated by the red arrows, is seen from a prolate to oblate shape when the monopole interaction is reduced.

This is a variation of the so-called type-II shell evolution, where significant changes in nucleon occupation numbers produce large shifts of effective single-particle energies. In the present case, the single-particle energies of the neutron $1i_{13/2}$ orbital and proton $1h_{9/2}$ orbital are effectively lowered, locating deformed prolate states at lower energy. The combined action of the monopole and quadrupole interactions thus allows for a near-degenerate coexistence of strongly- and weakly-deformed states in the region of the neutron-deficient mercury isotopes.

While this effect is present in all mercury isotopes around the neutron mid-shell, it is the small additional difference in pairing energy gain between odd-even and even-even nuclei, which tips the balance in favor of a strongly- or weakly-deformed ground state.

Type-II shell evolution plays an essential role in the quantum phase transition in zirconium isotopes and in the shape coexistence in nuclei around ^{68}Ni [105, 106]. It appears that type-II shell evolution also produces the abrupt odd-even staggering effect in mercury isotopes. As such, this insight links our results to features at different locations of the nuclear chart. This mechanism, that was qualitatively described in [5], is now put on quantitative grounds. Moreover, the relevant proton and neutron orbitals are identified. The MCSM calculations can now also be compared to other observables such as energy band structures and transition matrix elements from nuclear spectroscopy, Coulomb excitation and transfer reaction experiments.

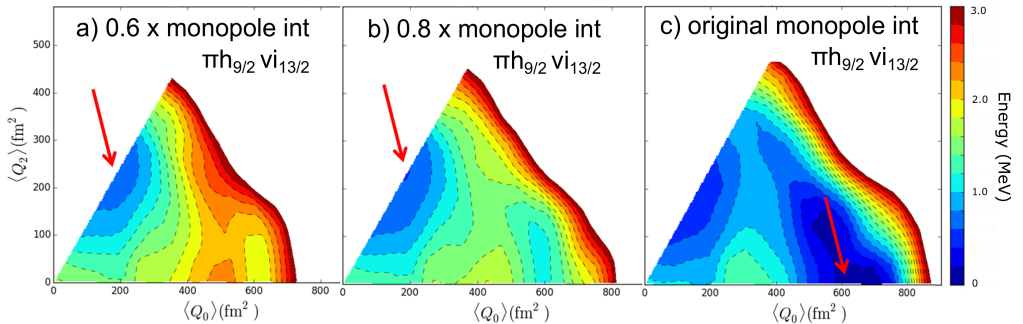


FIG. 18. Influence of the monopole interaction between $\pi h_{9/2}$ and $\nu i_{13/2}$ on the potential energy surface (PES) of ^{185}Hg . In panels A and B, the interaction is artificially reduced by a factor of 0.6 and 0.8 respectively with respect to the original shown in panel C. The red arrows indicate the PES minima.

V. Conclusion

We report on an experimental and theoretical study of the neutron-deficient mercury isotopes. By measuring for the first time the IS and HFS of $^{177-180}\text{Hg}$ and validating previous measurements along the chain of heavier mercury isotopes, the end-point of shape-staggering in the neutron-deficient direction was found to be at ^{180}Hg . Not only the $\delta\langle r^2 \rangle$ of $^{177-180}\text{Hg}$, but the magnetic and quadrupole moments of $^{177,179}\text{Hg}$ support the inference of their small deformation. More advanced descriptions were obtained from the MCSM and DFT calculations.

The DFT models offer an interpretation of the observed difference in charge radii in terms of the odd-even mass staggering combined with multiple near-degenerate minima of sufficiently different deformation. The UNEDF1^{SO} with modified pairing strength as well as the SLy5s1 and the SLy5s4 functionals provide a radius staggering that is similar to experiment, but predict a staggering pattern that sets in with two neutrons less. Reproducing all details of the observed staggering of the radii is not possible at the moment, as a quantitative description of the nuclear charge radii is extremely sensitive to many details of the functionals employed. For two such aspects, the pairing strength and the surface tension, the delicate balance between the minima and its consequence for the nuclear charge radii was shown explicitly.

The largest ever MCSM calculations were performed for this work. Both the magnetic and quadrupole moments and the radii changes calculated with the MCSM agree with the experimental results to a remarkable extent. These calculations point to the origin of the shape staggering as coming from a combination of pairing with a variation of type-II shell evolution. In the latter, the

monopole interaction between the $\nu i_{13/2}$ and $\pi h_{9/2}$ orbitals causes an enhanced occupation of these orbitals which drives the mercury isotopes to deformation. The change in their occupation from one isotope to the other causes their pronounced shape staggering. This understanding can now be tested in other areas of the nuclear chart, and the MCSM can be used to calculate different experimental observables.

VI. Acknowledgments

We would like to thank to ISOLDE collaboration and technical staff for providing excellent assistance during the experiment. This project has received funding from the European Union's Horizon 2020 research and innovation programme and the Seventh Framework Programme for Research and Technological Development under grant agreements 262010 (ENSAR), 267194 (CO-FUND), 289191 (LA³NET), 654002 (ERC-2011-AdG-291561-HELIOS). This work was supported by FWO-Vlaanderen (Belgium), by GOA/2010/010 (BOF KU Leuven) and by the IAP Belgian Science Policy (BriX network P7/12). This receive support from JSPS and FWO-Vlaanderen under the Japan-Belgium Research Cooperative Program. The MCSM calculations were performed on the K computer at RIKEN AICS (hp160211, hp170230). This work was also supported in part by Priority Issue on Post-K computer (Elucidation of the Fundamental Laws and Evolution of the Universe) from MEXT and JICFuS. This project was partially funded by a grant from the UK Science and Technology Facilities Council (STFC): Consolidated Grant ST/L005794/1. The DFT calculations with UNEDF1^{SO} were performed

using the DiRAC Data Analytic system at the University of Cambridge, operated by the University of Cambridge High Performance Computing Service on behalf of the STFC DiRAC HPC Facility (www.dirac.ac.uk). This equipment was funded by BIS National E-infrastructure capital grant (ST/K001590/1), STFC capital grants ST/H008861/1 and ST/H00887X/1, and STFC DiRAC Operations grant ST/K00333X/1. DiRAC is part of the National e-Infrastructure. DFT calculations with the SLy5sX parameterizations were performed at the Com-

puting Centre of the IN2P3 and at the Consortium des Équipements de Calcul Intensif (CÉCI), funded by the Fonds de la Recherche Scientifique de Belgique (F.R.S.-FNRS) under Grant No. 2.5020.11. S. S. acknowledges a SB PhD grant from the former Belgian Agency for Innovation by Science and Technology (IWT), now incorporated in FWO-Vlaanderen. L. P. G. acknowledges FWO-Vlaanderen (Belgium) as an FWO Pegasus Marie Curie Fellow. J.D. and A.P. acknowledge a partial support from the STFC grant No. ST/P003885/1.

-
- [1] J. Bonn *et al.*, Physics Letters B **38**, 308 (1972).
 [2] J. Bonn *et al.*, Zeitschrift für Physik A Atoms and Nuclei **276**, 203 (1976).
 [3] T. Kühl *et al.*, Phys. Rev. Lett. **39**, 180 (1977).
 [4] G. Ulm *et al.*, Zeitschrift für Physik A Atomic Nuclei **325**, 247 (1986).
 [5] K. Heyde and J. Wood, Review of Modern Physics **83**, 1467 (2011).
 [6] S. Frauendorf and V. V. Pashkevich, Physics Letters B **55**, 365 (1975).
 [7] N. Bree *et al.*, Phys. Rev. Lett. **112**, 162701 (2014).
 [8] L. P. Gaffney *et al.*, Physical Review C **89**, 024307 (2014).
 [9] K. Wrzosek-Lipska *et al.*, unpublished (2017).
 [10] R. Julin, K. Helariutta, and M. Muikku, J. Phys. G: Nucl. Part. Physics **27**, R109 (2001).
 [11] F. Kondev *et al.*, Physics Letters B **528**, 221 (2002).
 [12] A. Melerangi *et al.*, Phys. Rev. C **68**, 041301 (2003).
 [13] D. O'Donnell *et al.*, Phys. Rev. C **85**, 054315 (2012).
 [14] S. Schwarz *et al.*, Nuclear Physics A **693**, 533 (2001).
 [15] J. D. Cole *et al.*, Phys. Rev. C **30**, 1267 (1984).
 [16] D. G. Jenkins *et al.*, Phys. Rev. C **66**, 011301 (2002).
 [17] J. Elseviers *et al.*, Phys. Rev. C **84**, 034307 (2011).
 [18] J. Sauvage *et al.*, The European Physical Journal A **49**, 109 (2013).
 [19] E. Rapisarda *et al.*, Journal of Physics G: Nuclear and Particle Physics **44**, 074001 (2017).
 [20] A. N. Andreyev *et al.*, Phys. Rev. C **80**, 054322 (2009).
 [21] A. N. Andreyev *et al.*, Journal of Physics G: Nuclear and Particle Physics **37**, 035102 (2010).
 [22] R. Catherall *et al.*, Journal of Physics G: Nuclear and Particle Physics **44**, 094002 (2017).
 [23] V. Fedosseev *et al.*, Journal of Physics G: Nuclear and Particle Physics **44**, 084006 (2017).
 [24] T. Day Goodacre *et al.*, Nucl. Instrum. Methods B **376**, 39 (2016).
 [25] B. A. Marsh *et al.*, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **317**, 550 (2013), xVIth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 27, 2012 at Matsue, Japan.
 [26] B. Marsh *et al.*, Nature Physics, *Accepted* (2018).
 [27] T. Day Goodacre *et al.*, unpublished (2018).
 [28] T. Day Goodacre *et al.*, Hyperfine Interactions **238**, 41 (2017).
 [29] J. Bonn *et al.*, Zeitschrift für Physik A Atoms and Nuclei **272**, 375 (1975).
 [30] A. N. Andreyev *et al.*, Phys. Rev. Lett. **105**, 252502 (2010).
 [31] M. D. Seliverstov *et al.*, Phys. Rev. C **89**, 034323 (2014).
 [32] B. Lommel *et al.*, Nucl. Instrum. Methods A **480**, 199 (2002).
 [33] S. Kreim *et al.*, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **317**, 492 (2013), xVIth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 27, 2012 at Matsue, Japan.
 [34] R. N. Wolf *et al.*, International Journal of Mass Spectrometry **349**, 123 (2013).
 [35] F. Herfurth, J. Dilling, A. Kellerbauer, G. Bollen, S. Henry, H.-J. Kluge, E. Lamour, D. Lunney, R. Moore, C. Scheidenberger, S. Schwarz, G. Sikler, and J. Szerypo, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **469**, 254 (2001).
 [36] J. G. Cubiss *et al.*, Physical Review C **97**, 054327 (2018).
 [37] R. Brun and F. Rademakers, Proceedings AINHENP'96 Workshop, Lausanne, Nucl. Instrum. Methods A, <http://root.cern.ch> **389**, 81 (1997).
 [38] E. W. Otten, "Nuclear radii and moments of unstable isotopes," in *Treatise on Heavy Ion Science: Volume 8: Nuclei Far From Stability*, edited by D. A. Bromley (Springer US, Boston, MA, 1989) pp. 517–638.
 [39] W. Gins *et al.*, Computer Physics Communications **222**, 286 (2018).
 [40] G. Torbom, B. Fricke, and A. Rosén, Phys. Rev. A **31**, 2038 (1985).
 [41] B. Fricke and K. Heilig, Landolt-Bornstein **20** (2004), 10.1007/b87879.
 [42] D. Berdichevsky and F. Tondeur, Zeitschrift für Physik A Atoms and Nuclei **322**, 141 (1985).
 [43] R. J. Reimann and M. N. McDermott, Phys. Rev. C **7**, 2065 (1973).
 [44] O. Redi and H. H. Stroke, Phys. Rev. A **2**, 1135 (1970).
 [45] J. E. Rosenthal and G. Breit, Phys. Rev. **41**, 459 (1932).
 [46] A. Bohr and V. F. Weisskopf, Phys. Rev. **77**, 94 (1950).
 [47] J. Persson, Atomic Data and Nuclear Data Tables **99**, 62 (2013).
 [48] P. Moskowitz and M. Lombardi, Physics Letters B **46**, 334 (1973).
 [49] H. J. Rosenberg and H. H. Stroke, Phys. Rev. A **5**, 1992 (1972).
 [50] T. Fujita and A. Arima, Nuclear Physics A **254**, 513 (1975).

- [51] A. Mårtensson-Pendrill, Phys. Rev. Lett. **74**, 2184 (1995).
- [52] J. Bieron *et al.*, Phys. Rev. A **71**, 012502 (2005).
- [53] R. H. Kohler, Phys. Rev. **121**, 1104 (1961).
- [54] M. Anselment *et al.*, Nuclear Physics A **451**, 471 (1986).
- [55] S. B. Dutta *et al.*, Zeitschrift für Physik A Hadrons and Nuclei **341**, 39 (1991).
- [56] H. De Witte *et al.*, Phys. Rev. Lett. **98**, 112502 (2007).
- [57] M. D. Seliverstov *et al.*, The European Physical Journal A **41**, 315 (2009).
- [58] I. Angeli and K. Marinova, Atomic Data and Nuclear Data Tables **99**, 69 (2013).
- [59] M. P. Carpenter *et al.*, Phys. Rev. Lett. **78**, 3650 (1997).
- [60] F. G. Kondev *et al.*, Phys. Rev. C **62**, 044305 (2000).
- [61] F. G. Kondev *et al.*, Nuclear Physics A **682**, 487 (2001).
- [62] D. O'Donnell *et al.*, Phys. Rev. C **79**, 051304 (2009).
- [63] R. Bauer *et al.*, Nuclear Physics A **209**, 535 (1973).
- [64] N. Stone, Atomic Data and Nuclear Data Tables **90**, 75 (2005).
- [65] C. Ekström, H. Rubinsztein, and P. Möller, Physica Scripta **14**, 199 (1976).
- [66] A. E. Barzakh *et al.*, AIP Conference Proceedings **610**, 915 (2002).
- [67] A. E. Barzakh *et al.*, Phys. Rev. C **95**, 014324 (2017).
- [68] L. Bianco *et al.*, Physics Letters B **690**, 15 (2010).
- [69] I. Darby *et al.*, Physics Letters B **695**, 78 (2011).
- [70] T. Otsuka, T. Matsuo, and D. Abe, Phys. Rev. Lett. **97**, 162501 (2006).
- [71] A. de Shalit and I. Talmi, "Nuclear shell theory," (Academic Press, New York, 1963).
- [72] D. T. Yordanov *et al.*, Phys. Rev. Lett. **110**, 192501 (2013).
- [73] R. Ferrer *et al.*, Nature Communications **8**, 14520 (2017).
- [74] J. E. García-Ramos and K. Heyde, Phys. Rev. C **89**, 014306 (2014).
- [75] J. M. Yao, M. Bender, and P.-H. Heenen, Phys. Rev. C **87**, 034322 (2013).
- [76] S. Sakakihara and Y. Tanaka, Nuclear Phys. A **726**, 37 (2003).
- [77] J. M. Boillos and P. Sarriguren, Phys. Rev. C **91**, 034311 (2015).
- [78] V. Manea *et al.*, Phys. Rev. C **95**, 054322 (2017).
- [79] M. Stoitsov *et al.*, Computer Physics Communications **167**, 43 (2005).
- [80] M. Stoitsov *et al.*, Comput. Phys. Comm. **184**, 1592 (2013).
- [81] M. Kortelainen *et al.*, Phys. Rev. C **82**, 024313 (2010).
- [82] M. Kortelainen *et al.*, Phys. Rev. C **85**, 024304 (2012).
- [83] Y. Shi, J. Dobaczewski, and P. T. Greenlees, Phys. Rev. C **89**, 034309 (2014).
- [84] E. Chabanat *et al.*, Nuclear Physics A **635**, 231 (1998).
- [85] J. Bartel *et al.*, Nuclear Physics A **386**, 79 (1982).
- [86] N. V. Giai and H. Sagawa, Physics Letters B **106**, 379 (1981).
- [87] W. Ryssens, *Symmetry breaking in nuclear mean-field models*, Ph.D. thesis, Université Libre de Bruxelles (2016).
- [88] R. Jodon *et al.*, Phys. Rev. C **94**, 024335 (2016).
- [89] S. Krieger *et al.*, Nucl. Phys. A **517**, 275 (1990).
- [90] C. Rigollet *et al.*, Phys. Rev. C **59**, 3120 (1999).
- [91] A. Pastore *et al.*, Physica Scripta **2013**, 014014 (2013).
- [92] W. Ryssens, M. Bender, and P.-H. Heenen, (In preparation).
- [93] S. Noritaka *et al.*, Physica Scripta **92**, 063001 (2017).
- [94] "K-computer facility (official website): <http://www.aics.riken.jp/en/k-computer/about/>," .
- [95] B. A. Brown, Phys. Rev. Lett. **85**, 5300 (2000).
- [96] T. Otsuka *et al.*, Phys. Rev. Lett. **104**, 012501 (2010).
- [97] Y. Utsuno *et al.*, Phys. Rev. Lett. **114**, 032501 (2015).
- [98] A. Bohr and B. Mottelson, *Nuclear Structure*, Nuclear Structure No. v. 1 (World Scientific, 1998).
- [99] Y. Tsunoda *et al.*, Phys. Rev. C **89**, 031301 (2014).
- [100] Y. Utsuno *et al.*, Phys. Rev. Lett. **114**, 032501 (2015).
- [101] T. R. Rodríguez and J. L. Egido, Phys. Rev. C **81**, 064323 (2010).
- [102] B. Cheal and K. T. Flanagan, Journal of Physics G: Nuclear and Particle Physics **37**, 113101 (2010).
- [103] C. M. BAGLIN, Nuclear Data Sheets **99**, 1 (2003).
- [104] T. Otsuka and Y. Tsunoda, Journal of Physics G: Nuclear and Particle Physics **43**, 024009 (2016).
- [105] T. Togashi *et al.*, Phys. Rev. Lett. **117**, 172502 (2016).
- [106] S. Suchyta *et al.*, Phys. Rev. C **89**, 021301 (2014).

Part II

An ion-beam cooler and guide for IGLIS

Chapter 5

In-gas-jet laser ionization spectroscopy

The nuclear structure of the heaviest elements is governed by the interplay between the attractive strong and repulsive electromagnetic interactions [112]. Several models (Chapter 2), predict physics observables for the heavy elements [113, 114]. However, in many cases, firm experimental data to validate them is lacking. Determining spins, radii and moments of these heavy nuclei is mandatory to further develop the understanding of how atomic nuclei are built from their constituent protons and neutrons.

Because of the low production rates of the heavy elements, these experiments are very challenging and data is scarce, as can be seen in Figure 1. The current limit on the existence of the heavy elements is the recently named element oganesson ($Z=118$) [115, 116] while optical spectroscopy has been performed up to nobelium ($Z=102$) [19]. The HELIOS (Heavy Element Laser Ionization and Spectroscopy) project at KU Leuven, in which this thesis work is situated, aims to address the challenges that are currently hindering heavy-element laser-spectroscopic studies by developing a new laser-spectroscopy technique: In-gas-jet laser ionization spectroscopy.

5.1 In-gas-jet laser ionization and spectroscopy

The in-gas-jet laser ionization and spectroscopy technique builds upon the in-gas-laser ionization and spectroscopy technique (IGLIS), developed over the past 2-3 decades at KU Leuven [117–119]. The former technique can be categorized within the limits of the broader IGLIS technique as being a more specific variation. IGLIS was used at the recently decommissioned Leuven Isotope Separator On Line (LISOL) facility at Louvain-La-Neuve for both the production (as an ion source) and study (using laser spectroscopy) of radioactive ion beams (RIB). In the in-gas-cell laser ionization and spectroscopy technique, RIBs are stopped, neutralized and thermalized in a high-pressure gas cell (filled with ~ 300 mbar argon gas) at room temperature. A flow of noble gas leads the seeded atoms from the stopping point towards the exit hole of the cell. Before leaving the cell, resonant laser excitation and ionization of the isotopes takes place in the cell.

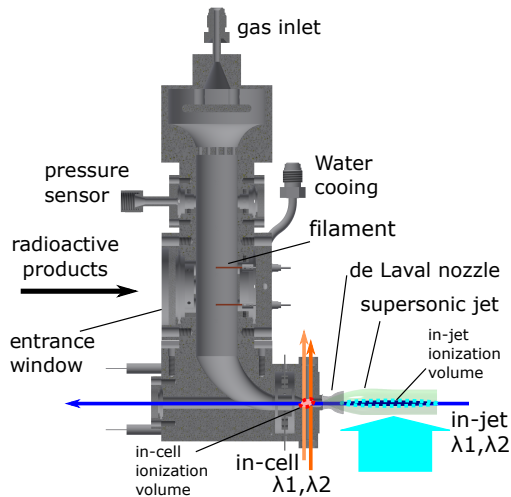


Figure 5.1: Cut view of the Leuven gas cell with the main components indicated. Figure adjusted from [120]

For in-jet spectroscopy, this process is very similar with the main difference being that the ionization region is moved from the gas cell to a supersonic gas jet. While different methods for the creation of a supersonic gas jet exist, a ‘de Laval’ nozzle was preferred for the setup at KU Leuven [85]. Such a supersonic jet has several advantages due to its low density, low temperature and high directionality when created under the right conditions. This technique combines

a high sensitivity with a high spectral resolution in comparison to other in-source spectroscopy methods such as e.g. the hot-cavity method used in Part 1 of this thesis to study mercury. In Figure 5.1, both techniques (in-cell vs in-jet) are shown and the main components of the cell are marked. The difference in position of the ionization volumes for in-cell and in-jet spectroscopy are also indicated.

The advantages of moving the ionization volume to the supersonic gas jet stem from the specific characteristics of a supersonic gas jet. As discussed in section 3.3, temperature and pressure play an important role in the spectral broadening of a measured hyperfine spectrum through Doppler and pressure broadening respectively. Equations 5.1-5.3 dictate the temperature (T), pressure P and density (ρ) in a supersonic jet of Mach number M . The Mach number is a dimensionless quantity that relates the flow velocity to the local speed of sound in a medium.

$$\frac{T}{T_0} = \left[1 + \left(\frac{\gamma - 1}{2} \right) M^2 \right]^{-1} \quad (5.1)$$

$$\frac{P}{P_0} = \left[1 + \left(\frac{\gamma - 1}{2} \right) M^2 \right]^{\frac{-\gamma}{\gamma - 1}} \quad (5.2)$$

$$\frac{\rho}{\rho_0} = \left[1 + \left(\frac{\gamma - 1}{2} \right) M^2 \right]^{\frac{-1}{\gamma - 1}} \quad (5.3)$$

Here, the subscript 0 denotes so-called stagnation values for P , T and ρ . These are the values inside the cell from which the supersonic jet propagates. The parameter γ refers to the ratio of specific heat capacities as $\gamma = C_p/C_v$ which is equal to 5/3 for mono-atomic gases. From equations 5.1-5.3, it is seen that P , T and ρ all scale inversely with M , resulting in reduced Doppler and pressure broadening (sec 3.3). For copper atoms, the FWHM of the Voigt profile of a spectral line drops with about 1 order of magnitude from ~ 2 GHz to ~ 200 MHz when considering a $T_0 = 300$ K, $P_0 = 100$ mbar gas cell with respect to a $M = 12$ supersonic jet [85]. Besides this increase in resolution, the first online test with the in-gas-jet technique at LISOL proved that this can even be achieved without a loss in efficiency [13]. Figure 5.2 shows the result of these first online tests from which the increase in spectral resolution is clear.

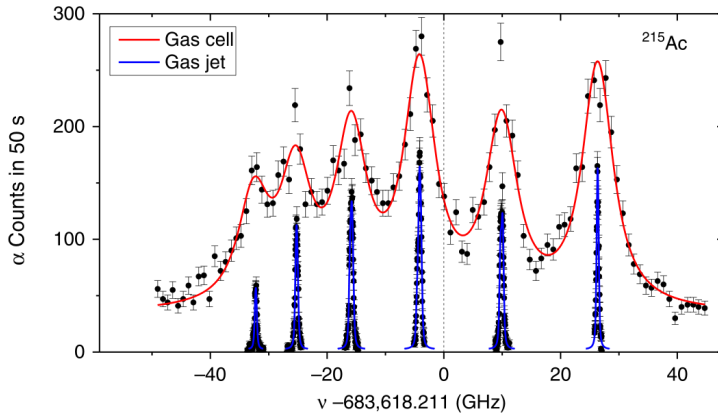


Figure 5.2: Comparison of in-gas-jet and in-gas-cell laser ionization spectroscopy of actinium isotopes studied at LISOL. Figure from [13]

When the cell is provided with a thin window through which an ion beam can enter and be stopped in a noble gas, this setup can be used both as an ion source and as a spectroscopy setup. If such a system is coupled to an in-flight facility, where the production of heavy-element RIB's is available, as is planned at SPIRAL2 in GANIL, it will be possible to perform laser spectroscopy on a broad scope of heavy elements which are currently out of reach [13].

5.1.1 Comparison to other laser spectroscopy techniques

Other well-established laser-spectroscopy experiments that aim to measure exotic nuclei can be categorized in either collinear or in-source type experiments. In the first type, lasers are overlapped with an isotopic beam in a collinear geometry. Making use of the reduced Doppler broadening of an accelerated ion beam, reaching the order of natural line widths for a 40 kV acceleration voltage, very high spectral resolution can be reached down to the order of ~ 10 MHz [121]. Here, both methods using photon detection of resonantly-excited atoms or ions and ion detection of resonantly-ionized atoms are well established at different facilities such as for instance by the COLLAPS and CRIS experiments at ISOLDE, CERN. However, even the more efficient CRIS technique can only reach isotopes with production rates in the order of 100 ions/s [121]. In-source ionization in either a hot-cavity or in a gas cell, is more efficient where, as shown in this work, isotopes with production rates down to 0.1 ions/s can be measured. Due to Doppler and pressure broadening however, in-

source techniques only provide spectral resolutions in the order of $\sim \text{GHz}$. This limits the applicability of the in-source methods to isotopes with large hyperfine splittings and isotope shifts. More specifically, medium to heavy-mass nuclei are often studied using this technique due to the increasing contribution of the Field shift ($\sim \delta\langle r^2 \rangle$) to the total IS [75]. In-gas-jet laser spectroscopy on the other hand could combine the high efficiency of conventional in-source techniques, with an improved spectral resolution by at least an order of magnitude. Using the fast stopping and extraction by a purified noble gas, isotopes with half-lives above 100 ms and produced at a rate of only 1 atom every 10 s can be measured with a spectral resolution of $\delta\nu/\nu \sim 1 \times 10^{-7}$ ($\sim 100 \text{ MHz}$) [13]. This makes the technique ideal for studying the heavy element region with high precision and efficiency. Since stopping and extraction of isotopes is in principle independent of chemical properties, the refractory elements can also be studied which are currently difficult to study with high-resolution spectroscopy at thick-target ISOL facilities.

5.2 Experimental setup at KU Leuven

At KU Leuven, a laboratory dedicated to the characterization and optimization of the in-gas-jet laser ionization technique has recently been commissioned. A schematic layout of the laboratory consisting of two experimental rooms is shown in Figure 5.3.

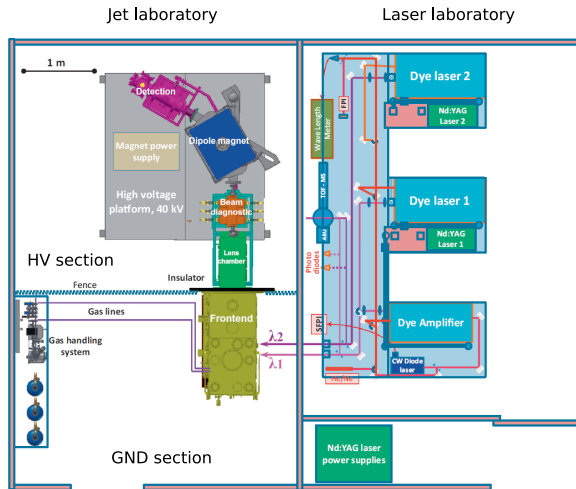


Figure 5.3: Layout of the IGLIS laboratories at KU Leuven. Figure from [120]

5.2.1 Jet laboratory

The jet laboratory, depicted on the left hand side of Figure 5.3, is divided in a high voltage (HV) and ground (GND) section, separated by an insulating fence. In the GND section of the lab, the frontend vacuum chamber, gas distribution and gas purification systems are placed. Inside the frontend vacuum chamber, the gas cell with a ‘de Laval’ nozzle for the production of the supersonic jets and radio-frequency quadrupole ion guides (RFQ) are situated. The RFQ’s are the main topic of the following chapters. The frontend is sectioned in three separate chambers for differential pumping named ‘jet chamber’, ‘DP1’ and ‘DP2’. This sectioning is needed to obtain a good-quality vacuum in the DP2 and beam-line sections to avoid electrical breakdown in the HV section and bad ion-beam emittances due to for instance reheating at extraction. The top of Figure 5.4 shows the layout of the inside of the frontend with the most important components indicated, while the bottom panel shows a picture of the actual setup.

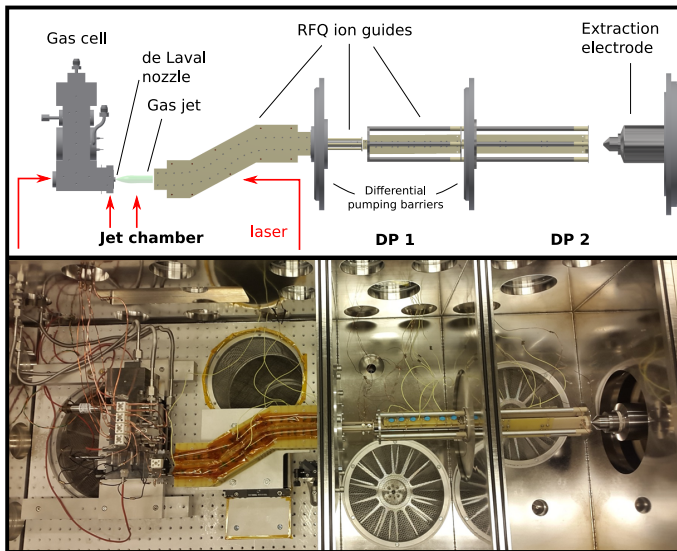


Figure 5.4: Top: Inside layout of the jet lab frontend at KU Leuven. Bottom: Picture of the actual setup.

In the HV section (Figure 5.5), the beam line connected to the frontend contains the lens chamber, housing an einzel lens and X- and Y-deflector plates. Behind the lens chamber, beam diagnostics are installed. These are a Faraday Cup

(FC) and a position-sensitive multi-channel plate (MCP) detector. A phosphor plate mounted at the back of the MCP is used to image the ion beam using a 90° prism and an Ethernet camera. A dipole bending magnet is used to mass separate the ion beam and send it to the focal plane of the beam line where a FC and MCP detector are installed. Time profiles of the ion beam can be measured with both MCP's, as explained in more detail in section 8.4.

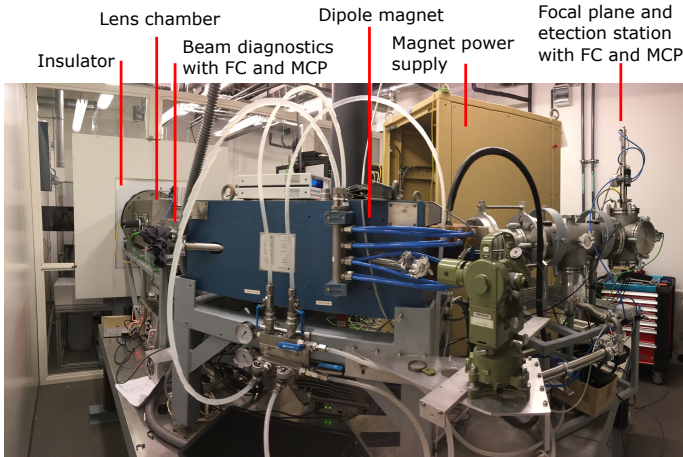


Figure 5.5: Picture of the HV section of the jet laboratory with the main components indicated.

5.2.2 Laser laboratory

The IGLIS laser laboratory shown in Figure 5.6 and described in [120] is situated in an ISO8 clean room containing the laser setup. The setup consists of two dye lasers (CREDO, Sirah Lasertechnik GmbH) and a two-stage dye amplifier being pumped by two Nd:YAG INNOSLAB lasers (EdgeWave GmbH). The latter have a maximum repetition rate of 15 kHz, which is necessary in order to irradiate all atoms in a 5 cm supersonic gas jet with ~ 500 m/s velocity at least once. These lasers have an output power of 90 W at 532 nm (frequency doubled) and 36 W at 355 nm (frequency tripled) with a 8-10 ns pulse length. Both dye lasers and the dye amplifier have a frequency-conversion unit (FCU) for frequency doubling and reaching the UV spectrum. The dye amplifier is seeded by a tunable single-mode CW laser (TA Pro, Toptica Photonics AG). The timing of the lasers is controlled by a digital-delay pulse generator (Quantum Composers) with several outputs for both triggering and delaying each laser separately. A home-made control program written by K. Dockx in XOJO is

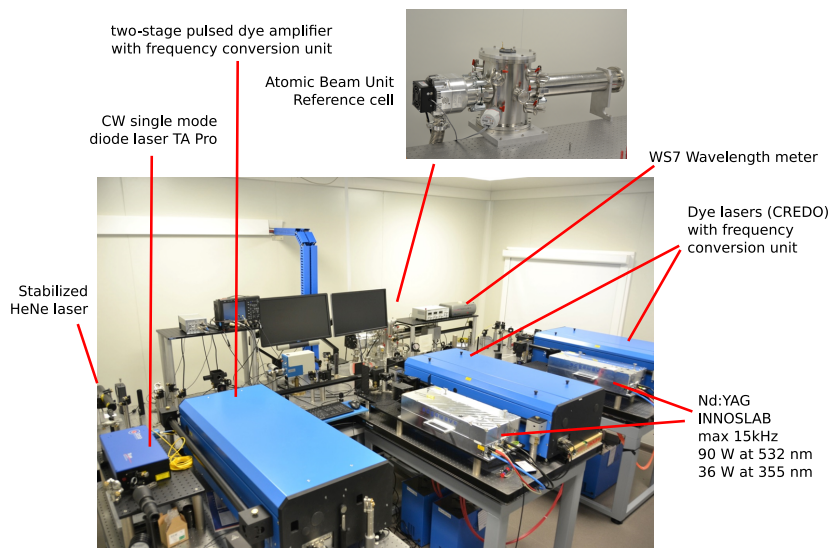


Figure 5.6: Picture of the laser laboratory clean room (ISO8) with most important components indicated.

used to control the lasers and automate laser scans with either the atomic beam unit (ABU) or the fluorescence-detection setup for gas-jet-formation measurements. This program is also used for stabilization of the CW laser based on a software PI-loop (partial PID-loop) using as input frequencies measured by a HF-ANGSTROM WS/7 (HighFinesse GmbH) wavelength meter. The input frequencies of the continuous-wave diode laser and stabilized Helium-Neon laser are simultaneously measured using an opto-mechanical switch (HighFinesse GmbH).

Chapter 6

Ion guide and beam cooler principles

Ion guides are devices for trapping, preparing and transporting charged particles. Beam coolers are a specific type of ion guides for optimizing ion-beam characteristics such as emittance and energy spread. In this chapter, the basic operational principles of such devices are described.

6.1 Ion trapping with RF potentials

The majority of ion guides make use of quadrupolar radio-frequency (RF) electric fields in order to confine the particles radially. The quadrupolar field is created by applying RF potentials to rods that are positioned in a configuration as shown on the left of Figure 6.1. By applying the potentials that are 180° out of phase to adjacent rods, at any moment in time, a potential is created that is confining in one direction but is anti-confining in the other. Such a potential is shown on the right of Figure 6.1. However, by fast switching of this saddle potential, a confining pseudopotential is created. Dehmelt proved this 50 years ago, by showing that an inhomogeneous RF field can create a non-zero time-averaged force F_n , which can be described as the gradient of a pseudopotential V_{pseudo} as $F_n = -e\nabla V_{\text{pseudo}}(r)$ [122].

The RF-fields of a linear RFQ only radially confine the charged particles in an ion guide. Additional fields are needed to confine or dictate the motion of the charged particles along the axis of the ion guide. This can be done using DC

potentials causing an axial field gradient that can be used for dragging the ions through the RFQ or even for temporarily collecting and storing the ions as is for instance done in ion bunchers.

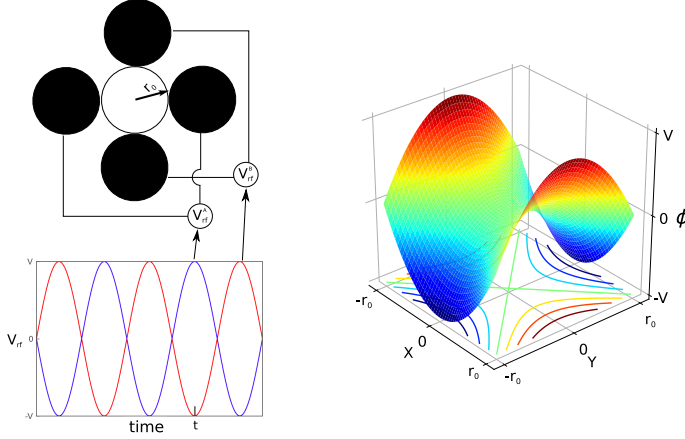


Figure 6.1: Left top: Rod configuration for producing a radially confining electric field using RF potentials. Left bottom: Time dependence and relative phase of the applied potentials. Right: Representation of the saddle potential created by applying potentials 180° out of phase to adjacent rods at time t .

Equations of motion

The oscillation of an ion in the RF field gives rise to two types of motion: the micro motion, an oscillation with the frequency of the driving RF field and the macro-motion that slowly drives the ion as a harmonic oscillator (see Figure 6.2). As RFQ's make use of a confining quadrupole potential, the magnitude of the electric field strength in the radial direction increases linearly with the distance from the trap center. The same is true for the magnitude of the instantaneous force $F = eE$ acting on the ions and also the time-averaged force F_n . The ion will therefore perform a bound, harmonic (macro) motion about the trap center [123]. In linear RFQ's the voltage

$$V(t) = V_{DC} + V_{rf} \sin(\omega_{rf} t) \quad (6.1)$$

applied to the electrodes, gives rise to a pseudopotential well that has the form of a parabola, described by

$$V_{\text{pseudo}}(r) = \frac{eV_{rf}^2}{4mr_0^4\omega_{rf}^2} r^2. \quad (6.2)$$

Here, r_0 is the radius of the inscribed circle of the RFQ electrodes, referred to as the characteristic distance, m is ion mass, e is the ion charge, ω_{rf} is the RF angular frequency and V_{rf} is the peak-to-peak RF amplitude.

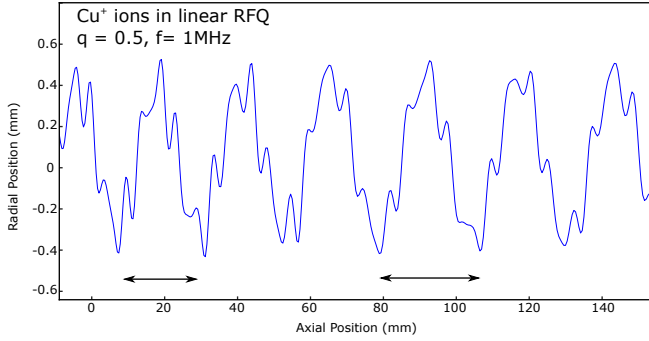


Figure 6.2: Simulated motion of Cu^+ ions in a linear RFQ showing the large-amplitude macromotion and small-amplitude micromotion superimposed. The effect of the axial DC dragging field is seen as the increase in axial movement per cycle (indicated by the black arrows).

Stable ion motion is not valid for random combinations of variables. The equation of motion in fact dictates what parameter set results in a stable ion motion.

The quadrupole potential has the form of

$$\phi = \frac{\phi_0(x^2 - y^2)}{2r_0^2} \quad (6.3)$$

where ϕ_0 refers to the peak-to-peak voltage applied to the rods as given in 6.1. When focussing on the x dimension, and using $F_x = -e\nabla\phi_0(y=0)$, the equations of motion become

$$m \frac{d^2x}{dt^2} = \frac{-2e(V_{DC} + V_{rf} \cos(\omega_{rf}t))x}{r_0^2} \quad (6.4)$$

As it turns out, there is no simple analytic solution for these equations but they can be transformed into the form of the so-called Mathieu equation, by rearranging the terms in equation 6.4.

$$\frac{d^2x}{d\xi^2} + (a_x - 2q_x \cos 2\xi)x = 0 \quad (6.5)$$

where ξ is related to the frequency as $\xi = \omega_{rf}t/2$, while a and q are referred to as the Mathieu parameters given by equations 6.6 and 6.7. The derivation is exactly similar for the y-dimension, but due to the 180° phase difference, a and q differ in sign.

$$q_x = -q_y = \frac{2eV_{rf}}{mr_0^2\omega_{rf}^2} \quad (6.6)$$

$$a_x = -a_y = \frac{4eV_{DC}}{mr_0^2\omega_{rf}^2} \quad (6.7)$$

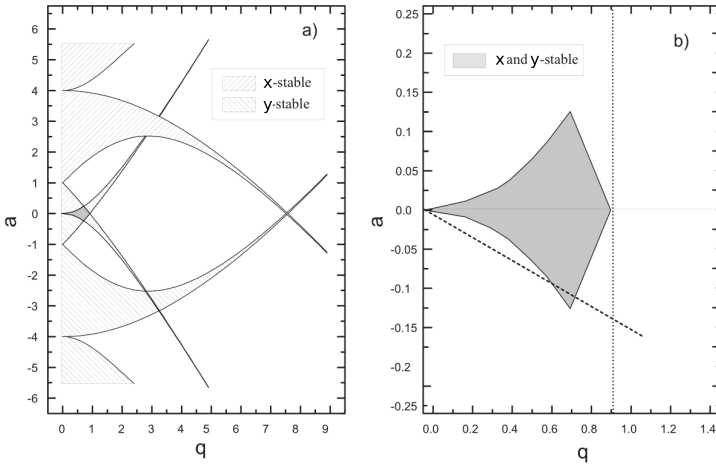


Figure 6.3: Mathieu stability diagram for ion motion as a function of the stability parameters a, q given in equations 6.6 and 6.7. Figure from [123].

It should be noted however, that U_{DC} here, does not refer to the axial DC potential that is applied for instance as a DC gradient in ion guides to guide ions through the device, but rather is a fixed potential applied to the rods as given in 6.1. In our case, when working with an RF-only linear RFQ, equation 6.5 is reduced as $a = 0$. In this specific case, the parameter region where the ion motion becomes stable is given by $0 < q < q_{\max} = 0.908$. Another remark deals with the limitation of the Mathieu stability diagram. Since this diagram treats only two types of ion trajectories, bound trajectories and unbound trajectories, the physical constraints imposed by the finite size of real quadrupoles are not considered [124]. Simulations of the RFQ's are needed to fully characterize the system in advance and should be experimentally verified.

6.2 Buffer gas cooling

Buffer-gas cooling of ion beams is a well-established method of reducing the emittance, and energy spread of an ion beam [125]. Emittance is a measure of the quality of ion beams related to the average spread of particle coordinates in both position and momentum phase space. Using collisions of trapped ions with a low-pressure gas, usually referred to as a “buffer gas”, ion beams can be cooled to close to the temperature of the gas itself. Buffer-gas damping of ion motion in a trap was first demonstrated in 1965 for mercury ions in a Paul trap into which a small amount of helium was introduced [125]. At high gas pressures and low ion energies, the effect of collisions with gas molecules is most often modelled as a viscous drag force that damps the oscillation of the ion in RF fields as $F_d = \frac{e}{K}v$. A measure of the strength of this effect is the ion mobility K , which is characteristic of a certain ion and buffer-gas combination. As the damping effect is proportional to the number of ion-gas collisions, reduced mobilities K_0 are most often quoted for a gas density of an ideal gas at 0°C and 1 atm and need to be rescaled for temperature and pressure. As a result of this damping force, the ion velocity is said to reduce exponentially according to

$$v = v_0 e^{\frac{e}{mK}t} = v_0 e^{\frac{e}{mK_0} \frac{N}{N_0} t}. \quad (6.8)$$

This model is often used in simulations of buffer-gas filled RFQ’s. However, this simple model breaks down microscopically and can for instance not model RF heating, discussed later, or the random statistical nature of ion-gas collisions and can significantly overestimate the effectiveness of the cooling process [126].

Another often-used model for ion-gas interactions in simulations is the hard-sphere model, which, as the name suggests, models the collisions as colliding hard spheres. In this model, the ions scatter on gas particles with a collision frequency related to the ion- and gas-particle’s diameters, gas pressure and velocities. The gas particles travel in randomized velocities according to the Maxwell-Boltzmann distribution. This is a more microscopic approach to modelling the buffer gas effect, but is computationally more intensive than the viscous drag model. However, modelling the full interaction potential as a hard-sphere potential oversimplifies interactions at larger distances.

As detailed in [125,126], the actual form of the ion-molecule interaction potential depends on the atomic structure of the ion and the molecule. The attractive dipole-induced interaction, has a $1/r^4$ dependence, and dominates the interaction potential at larger distances. Similarly, an induced quadrupole effect with an attractive $1/r^6$ dependence is present. Higher-order terms associated with a strong repulsive core, can be approximated by a single higher-order positive term with a $1/r^n$ dependence. The resulting ion-molecule interaction potential

is modelled as

$$V(r) = \frac{B}{r^n} - \frac{C_4}{r^4} - \frac{C_6}{r^6} \quad (6.9)$$

where B , C_4 and C_6 are constants depending even on the electronic structure of the ion and molecule. While other parametrizations than the so-called (n,4,6) potential in equation 6.9 are used in parallel, rather simple relations exist to convert one to the other [125,126]. When the full interaction potential is known, the ion-gas collisions can be modelled in detail as is for instance implemented in the IonCool software, developed at MSU [126]. A comparison of several models is given in Ref. [127], which states that ‘if one is interested in a fast and possibly more qualitative evaluation of a problem, the hard-sphere model will suffice. However, for quantitative results and comparisons with experimental results the realistic model is recommended’.

RF heating

An important constraint on the applicability of buffer-gas cooling is due to the molecular mass of the buffer gas. In RF-driven coolers, the so-called RF-heating of ions becomes important when the mass of the buffer-gas molecule M is comparable or larger than the mass of the ion m .

If there is only little momentum transferred in ion-gas collisions, there is an average cooling effect. For maximum momentum transfer, however, the collisions lead to a larger paths in the RFQ. For $M = m$ for example, the ion is at rest immediately after the collision, but then starts out on a larger orbit in phase space. In this case the ion picks up energy from the RF field. As noted in [127], the average energy transfer per hard-sphere collision can be calculated as a function of the mass ratio $\kappa = M/m$ as

$$\langle \varepsilon_{RF} \rangle = \kappa \frac{\kappa - 1}{(1 + \kappa)^2}. \quad (6.10)$$

For values $0 < \kappa < 1$ this expression is always negative, so a net cooling effect is present. For $\kappa > 1$, ε_{RF} is positive and on average energy is added rather than dissipated in collisions. This is why, helium is used mostly as a buffer gas in RFQ’s. While cooling times are slightly larger for lower-mass buffer gases, they can cool more ion species. Buffer gases are almost always noble gases as they are chemically less reactive which reduces loss of ion species due to molecule formation. In addition, the high ionization potential of helium or argon ($\text{IP}(\text{He}) = 26,4 \text{ eV}$, $\text{IP}(\text{Ar}) = 15,8 \text{ eV}$) reduces the possibility of charge exchange. The effect on the temperature due to RF fields are not easily estimated. In general, it has to be determined by experiment or by Monte-Carlo simulations of the ion-molecule interaction. [125]

Chapter 7

Simulations and design

The goal of the ion guides in the IGLIS setup at KU Leuven can be divided in several parts. Firstly, they have to efficiently capture the ions created in the supersonic gas jet or in the gas cell. Secondly, the ions need to be transported from the high-pressure jet-chamber region to the beam line which is under a high-vacuum, high-voltage regime (see Figure 7.1). The ion guides have to do this while providing optimal transport efficiency, which is crucial when working with exotic nuclei, as well as a good beam quality. In order to design the RFQ ion guides, all these aspects have to be taken into account. The most important factors of the design of the RFQ system for the IGLIS studies at KU Leuven are described in this chapter. By doing simulations with different programs, the operational characteristics were calculated and optimized and later on compared to the results of commissioning experiments described in Chapter 8.

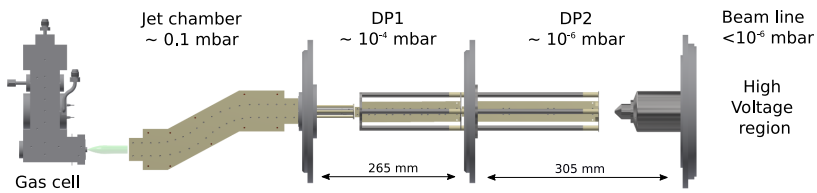


Figure 7.1: Frontend design with different pressure regimes.

7.1 Design

The atoms of interest are extracted in an argon gas jet expanding from the buffer-gas cell. For the gas jet to remain stable, and not to over- or under expand, the background pressure in the region of the supersonic jet has to match the pressure of the jet itself. For a jet with Mach number $M = 8$ and a gas cell pressure of $P_0 = 300$ mbar, this would impose the pressure in the whole jet chamber to be ≈ 0.1 mbar according to Equations 5.1-5.3 [85]. At roughly 0.5 m from the jet chamber, the laser-produced ions will be accelerated by a high voltage (HV) and sent to a dipole mass-separating magnet. Such a region of high voltage (up to 40 kV) requires a vacuum of the order of 10^{-6} mbar to prevent electrical breakdown in the HV region. This means that the ions have to be transported from a high-pressure (≈ 0.1 mbar) to a low-pressure region ($\approx 10^{-6}$ mbar). This requirement is met by introducing two stages of differential pumping and therefore also three different RFQ structures named S-shaped RFQ (SRFQ), differential pumping RFQ (DPRFQ) and ion guide RFQ (IGRFQ). The sizes of the pumping diaphragms (11 mm diameter) were based on vacuum calculations that included both the inflow of gas from the gas cell and the pumping system. For RFQ simulation purposes, these vacuum calculations were also used to estimate the pressures in chambers DP1 and DP2, when the pressure in the jet chamber is kept fixed, as dictated by the jet Mach number and stagnation pressure.

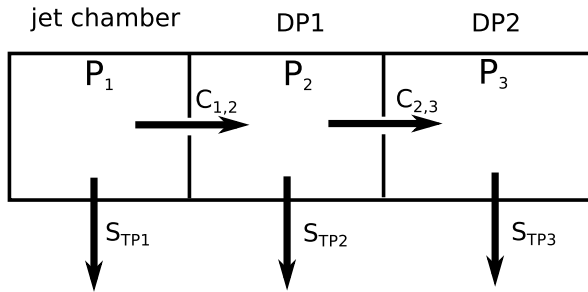


Figure 7.2: Vacuum calculations diagram.

Using the diagram of the vacuum calculations as shown in Figure 7.2, the pressures in DP1 and DP2 (referred to as P_2 and P_3 respectively), were calculated as

$$P_2 = P_1 \frac{C_{1,2}}{S_{TP2}} \quad P_3 = P_2 \frac{C_{2,3}}{S_{TP3}} \quad (7.1)$$

The conductances were estimated using formulas from [128] as

$$C_{\text{tube}} = \frac{\pi}{12} \bar{v} \frac{d^3}{l} \quad \text{with} \quad \bar{v} = \sqrt{\frac{8kT}{\pi M}} \quad (7.2)$$

$$C_{\text{app}} = A \sqrt{\frac{RT}{2\pi M}}. \quad (7.3)$$

where d , l , k , T , M , A and R represent the diameter, length, Boltzmann constant, temperature and molar mass, surface area and gas constant respectively.

7.1.1 SRFQ

Working with a unidirectional supersonic gas jet, it is important to consider that the gas jet does not enter directly the differential pumping diaphragm separating two chambers. Doing so renders the differential pumping invalid. By bending the RFQ, such as for example by 90° or in an S-shape, the gas jet can be directed away from the pumping diaphragm.

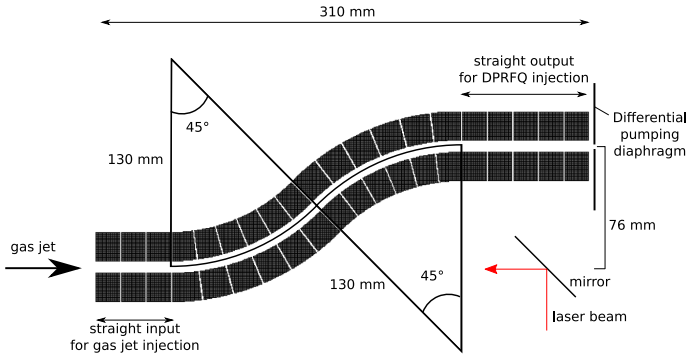


Figure 7.3: Design of the SRFQ.

The S-bend is designed such that it is made up of two soft 45° bends with a bending radius of 130 mm from the RFQ center, each bending in opposite direction to each other as shown in Figure 7.3. Ensuring a soft bending increases the transmission through a bent RFQ. This can be conceptually understood by thinking of ions moving through the RFQ as marbles going down a slide. If the axial energy of the ions is high enough to overcome the pseudo-potential well depth, ions might not make it through the curved section. The magnitude

of the bend however also allows for an easy positioning of a mirror to send a laser beam in counter propagation to the gas jet. In order to have an optimal injection and extraction from the SRFQ, the entrance and exit sections were designed as regular straight RFQ sections. The RFQ is segmented in 20 sections of electrodes to which an axial DC potential gradient can be applied to guide the ions along the RFQ's longitudinal axis.

When considering the size of the SRFQ, the gas jet plays an important role. In order to not disturb the gas-jet formation, the RFQ inner radius was chosen as such that even high-Mach-number jets of up to $M = 15$ can enter the RFQ (see Figure 7.4).

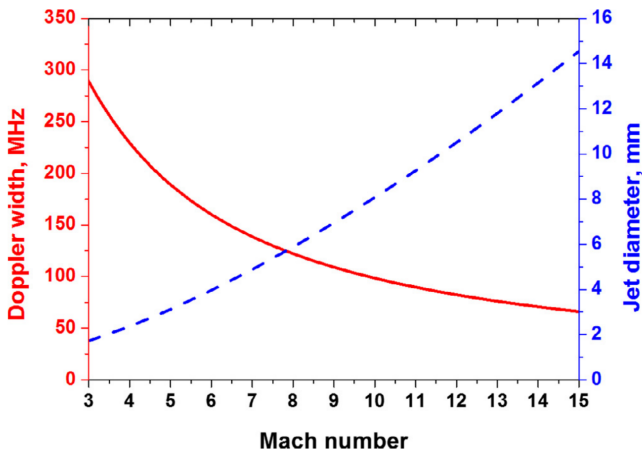


Figure 7.4: Relationship between jet diameter and Doppler width for supersonic jets of different Mach numbers. Figure taken from [120]

This avoids the formation of turbulences and, as such, a potential reduction of spectral band width of laser-spectroscopic studies. It also makes a proper injection into the RFQ easier. Therefore, r_0 was chosen as 9 mm. The final and constructed design of the SRFQ is shown in Figure 7.5.

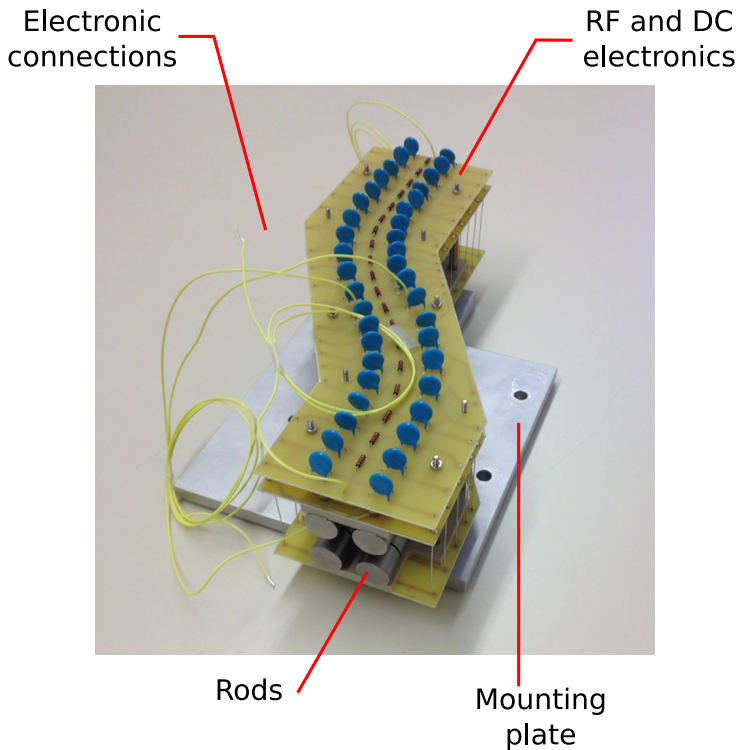


Figure 7.5: Photograph of the SRFQ ion guide.

7.1.2 DPRFQ

The DPRFQ is designed for enhanced differential pumping and therefore resembles a small, tubular section. It was inspired by a similar RFQ designed at MSU and presented in [129, 130]. The rods are cut in such a way that they take up minimal volume, but still produce a well-defined confining quadrupole potential. As the device is small, segmentation into miniature RFQ segments would make the construction complex. Therefore, the axial DC field is not provided by applying different DC voltages to several segments, but rather by designing two wedge-shaped electrodes only carrying a DC potential. These wedges are cut, such that when combined, they form a tube surrounding the RF-carrying rods. Wedge 1 has the bulk of its material at the entrance of the DPRFQ and carries a lower DC voltage than Wedge 2 and vice versa.

The radius of the rods was varied in a simulation in order to optimize transmission for a minimal size and maximum differential pumping (Figure 7.6). The final radius of the DPRFQ was chosen as $r_0 = 3$ mm. This allowed for a near 100% transmission, whilst keeping an 11 mm diameter hole in the differential pumping diaphragm. The tubular shape of the RFQ reduced its molecular-flow conductance by a factor 7 when modelled as an ideal tube with respect to a regular aperture.

In order to optimize the transition from the SRFQ to the DPRFQ, the RF rods of the DPRFQ extend through the differential pumping diaphragm and enter inside the SRFQ for ~ 3 mm. A picture of the final design is shown in Figure 7.7. However, the mounting flange in this photograph is mounted in reverse position for better visibility of the RFQ structure.

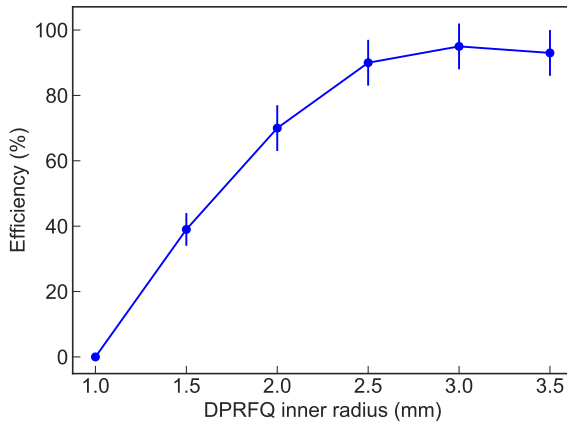


Figure 7.6: Transmission efficiency of the DPRFQ versus inner radius r_0 .

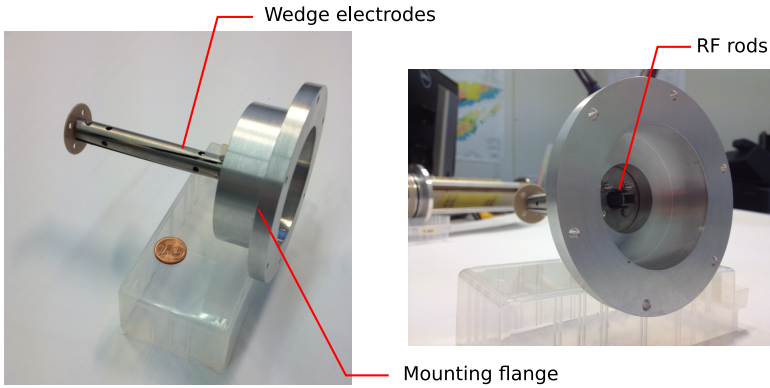


Figure 7.7: Picture of the DPRFQ ion guide.

7.1.3 IGRFQ

The last of the three RFQ structures has two main purposes: To guide the ions from the DP-RFQ up to the high-voltage extraction electrode, which will lead the ions to the mass-separation region. During this journey, the ions cross the boundary separating the DP-1 and DP-2 chambers through a narrow enough pumping orifice that allows for the vacuum constraints to be kept. This RFQ has an inner diameter of $r_0 = 4$ mm and is split in two by a pumping diaphragm with a hole of 11 mm diameter and 1 mm thickness. The RFQ is divided in 8 segments, 5 of which are situated before the diaphragm and 3 behind.

The final section of the RFQ is close to the extraction electrode which is kept at a voltage of -2000 V. In order to minimise the penetration of the HV field from the electrode into the IGRFQ, an extraction plate is placed as the final electrode on which only a DC voltage is applied (see Fig. 7.8). The shape, hole size and position of this electrode were simulated in order to optimize transport efficiency and minimise the field penetration. Based on these simulations, a straight plate with a thickness of 1 mm and a 4 mm hole was chosen.

Having the RFQ extraction in a low-pressure region when the ion beam is already cooled in a previous stage removes the possibility of reheating. This reheating process would be caused by the scattering of ions exiting the RFQ with remnant gas particles. Without the presence of a confining force, such as those provided by the RFQs, this would lead to a larger beam emittance.

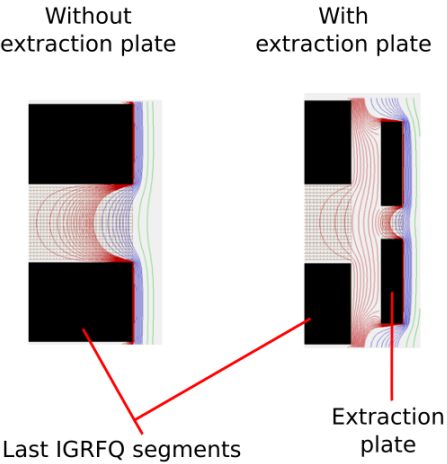


Figure 7.8: Simulated effect of an extraction plate on the penetration of the electric field inside the final IGRFQ section. Contour lines differ 1, 10 and 50 V for consecutive red, blue and green lines respectively. The simulations shown here were performed with SIMION (Sec. 7.2).

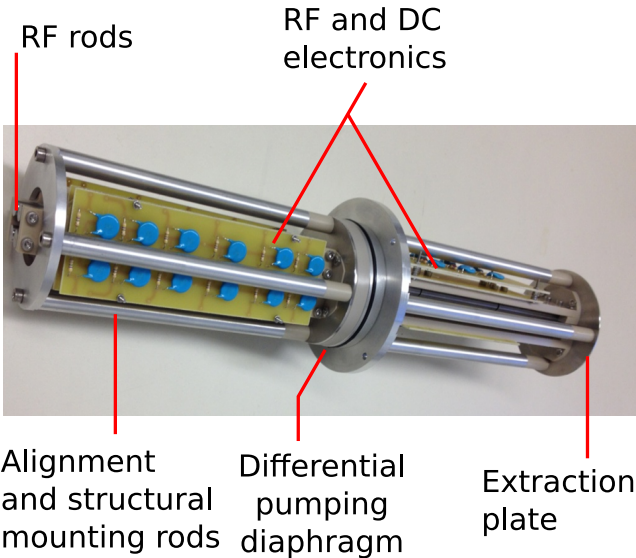


Figure 7.9: Photograph of the IGRFQ ion guide.

7.1.4 Complete Setup

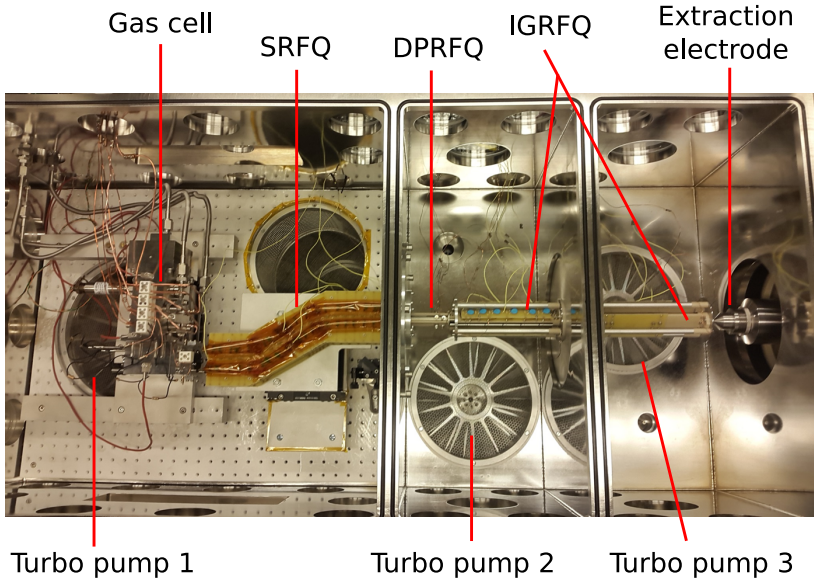


Figure 7.10: Photograph of the complete setup of ion guides installed in the IGLIS frontend vacuum chamber.

Figure 7.10 shows all three RFQ's installed in the IGLIS frontend vacuum chamber. The finalized RFQ designs were simulated in SIMION together with the beam line and dipole magnet.

7.2 SIMION and IonCool simulations

In order to investigate the performance characteristics of the device, simulations were performed. The electric fields created by the RFQ's were calculated with SIMION 8.0 [131], a standard ion optics software package for solving the Laplace equations for a given user-defined electrode configuration. Boundary conditions for solving these equations are defined by the electrode configuration and the voltages applied to them. Using user-written programs, it is possible to simulate ion trajectories moving through the combination of RF and DC fields as obtained from the electrode configuration. The effect of ions moving through a buffer gas can be included in such user programs. The hard-sphere model HS1 [132] is

for instance included in SIMION examples and can be implemented in a users' own simulation. Electric fields calculated by SIMION can also be extracted and used in other software. The IonCool software package [126] for instance, makes use of this feature to calculate ion trajectories and distributions from SIMION's electric-field calculations. However, IonCool provides the extra possibility to simulate ions moving through a buffer gas, using the so-called 'realistic potential' ion-buffer gas collision model. This model, detailed in e.g. [125–127], based on the $(n,4,6)$ parametrization of the ion-buffer gas interaction from Section 6.2, takes both short and long-range interactions into account. In this work, simulations of the ion trajectories were performed using both SIMION and IonCool software packages. From these trajectories, other operational characteristics such as transport efficiency, flight time or emittance can then be extracted.

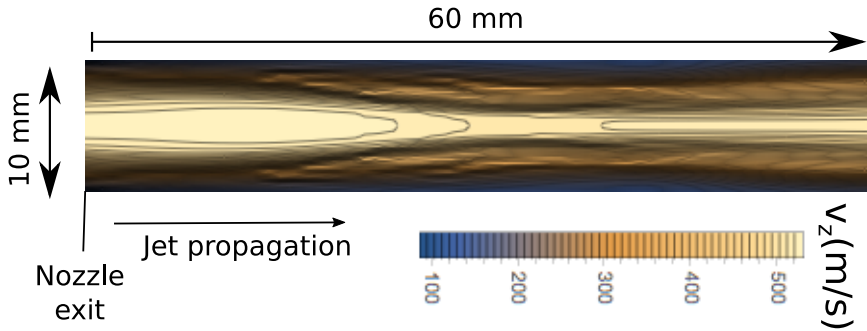


Figure 7.11: Longitudinal velocity profile of Mach 8 jet as simulated by COMSOL. COMSOL calculations provided by E. Mogylevsky.

As a starting point for all simulations, the initial conditions have to be defined. In this case, the initial spacial and velocity distribution of ions were provided by COMSOL simulations of a supersonic gas jet expanding from a de Laval nozzle with 1 mm throat diameter and 1 cm final outer radius. COMSOL Multiphysics [133] is a wide-range simulation software consisting of several different options such as computational fluid dynamics (CFD), and RF and DC electrical field. The simulations providing the ions initial conditions were performed by E. Mogylevsky using the COMSOL CFD module (see [134] for more info). The velocity distribution that was used as input is shown in Figure 7.11. Copper ions were used as test cases, as this element would also be used to characterize the setup experimentally. With an atomic mass of $A = 63, 65$, copper is sensitive to RF heating due to its similarity with argon ($A = 40$). Heavier atoms are expected to be cooled more efficiently.

Three different ion-buffer gas interaction models, explained in Section 6.2, were used to simulate the devices performance and results of the simulations are compared to experiment in Chapter 8. Atomic and ionic radii needed for the ion-buffer gas drag-force and hard-sphere models were obtained from [135]. The Cu^+ -Ar potential from [136] is shown in Figure 7.12 and was used as an input for the realistic potential model calculations. It is shown together with Hg^+ -Ar and Ar^+ -He interaction potentials used in [126], as a reference for typical deviations.

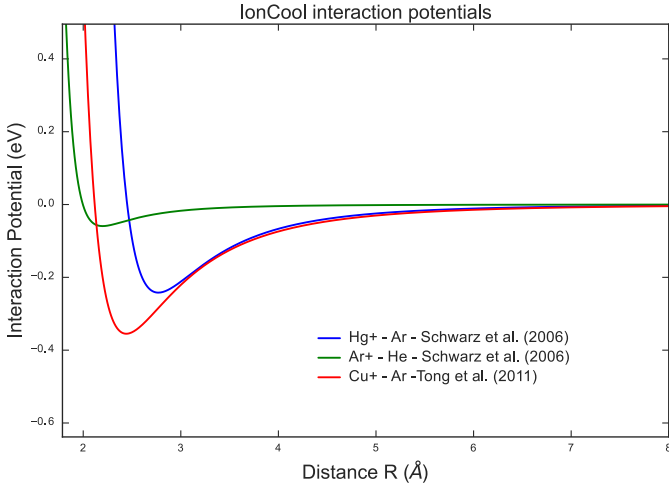


Figure 7.12: Interaction potentials for ion-buffer gas collisions.

7.3 Electronics

The RF voltages for operating all three RFQ's are generated using resonance circuits according to the layout shown in Figure 7.13. The sinusoidal voltages are created by TTI TG2511 function generators with a maximum output peak-to-peak voltage of $10 V_{pp}$. The amplitude of the RF applied to the RFQ's is regulated by the output voltage of these generators. The function generators of all three RFQ's are synchronized and locked in frequency and phase. Total transmission measurements show that phase- and frequency matching is necessary to operate under optimal conditions. The RF voltages are passed on to an RF amplifier (HLA150 plus HF linear amplifier) with a maximum output power of 80 W. This voltage is sent through a passive standing-wave-ratio (SWR) meter that indicates the ratio of outgoing versus reflected power and

provides a measurement of how well the impedances of the RF amplifier and the RFQ's acting as load are matched. The amplified RF signal is fed into a homemade impedance matched resonance circuit, as shown in Figure 7.14.

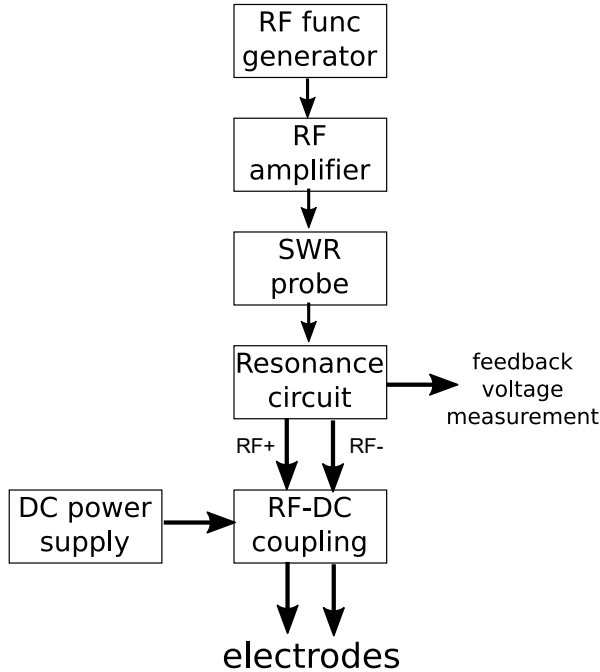


Figure 7.13: Layout of RFQ electronics.

The impedance is matched using an RF ferrite core transformer with one primary and two secondary coils. The secondary coils are wound in a counter-propagating direction and carry voltages that are 180° out of phase, labelled RF^+ and RF^- . Inductor coils were wound around PVC cylinders to add more inductance to the circuit and tune the resonance frequency to 1 MHz. A tunable capacitor is used to fine-tune the resonance. For the SRFQ, additional inductor coils were made with more windings to bring the resonance frequency down to 500 kHz. This latter option could be necessary when ions have to be guided using voltages higher than the gas-discharge breakdown limit. According to Equations 6.6 and 6.7, the voltages required to operate at a certain stability parameter would be lower by a factor 4 but would result in a shallower pseudopotential (Equation 6.2). For all RFQ circuits, one of the inductor coils carries a secondary winding such that it acts as a step down transformer. This smaller signal is used as a feedback to measure the RF that is applied to the rods. It is fed into homemade

rectifier circuit that is based on a full-wave diode bridge, which outputs a DC signal that is directly proportional to the AC input signal.

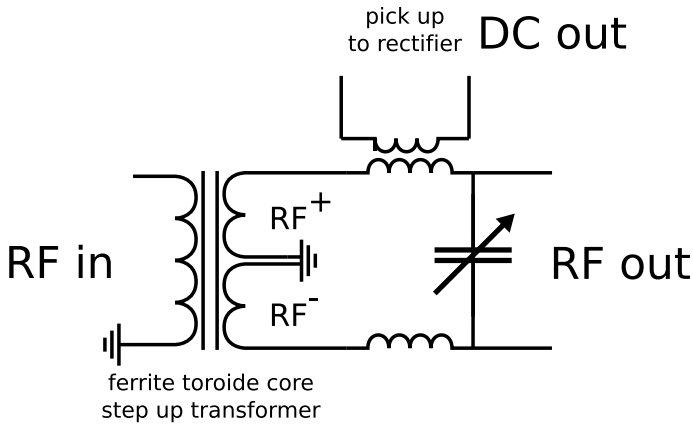


Figure 7.14: Resonance circuit for generation of the RF voltages.

The two out-of-phase output RF signals are combined with the DC voltages before finally being sent to the electrodes. For the SRFQ and IGRFQ, the RF and DC combination happens on a printed circuit board mounted on top of the RFQ. Here, besides the coupling of RF and DC, a DC voltage gradient that drags the ions through the RFQ's is created by a resistor chain. In case of the SRFQ for instance, this chain makes sure that the DC voltage on each of the 20 segments, lies 1 step lower by 1/20th of the total voltage gradient. The DPRFQ quadrupole rods carry a DC offset voltage induced by placing a voltage supply in between the transformer secondary windings and ground. The voltage gradient is created by the surrounding wedge electrodes.

A software program was written in XOJO by Kristof Dockx that allows the RF voltage amplitudes to be scanned for all RFQ's with respect to the Faraday cup signal recorded with a Keithley 6485 picoammeter to find the voltages for maximal transmission.

Chapter 8

Commissioning tests

The operation of the RFQ's has been characterized by performing commissioning tests of the total transmission efficiency, time profiles, emittance and longitudinal energy measurements. These tests have been confronted with the simulations detailed in Chapter 7. In order to obtain the working regime in which the SRFQ can be operated with regard to electrical breakdown, the Pashen curve of the SRFQ has been measured. To finalize the commissioning of the IGLIS laser- and jet laboratories, mass scans have been performed with the test case of laser-ionized copper isotopes.

8.1 Discharge tests

The operational regime at which the RFQ's can work in a gaseous environment can be strongly restricted by electrical discharges. Free ions or electrons that are accelerated in an electric field cause an avalanche effect by collisional ionization of gas molecules which are then accelerated themselves. When the breakdown voltage ' V_{br} ' is exceeded, which depends on the electrode distance, gas type and pressure, a discharge or electric arc can be formed. Pashen's law describes the breakdown voltage as a function of the product of pressure and characteristic electrode distance pd [137]. An example of such curves for different common gas types is shown in Figure 8.1.

Pashen-curve measurements are usually performed in well-controlled parallel-plate experiments [139]. In a complex system, where voltage differences are applied to electrodes and wires with differing characteristic distances d ,

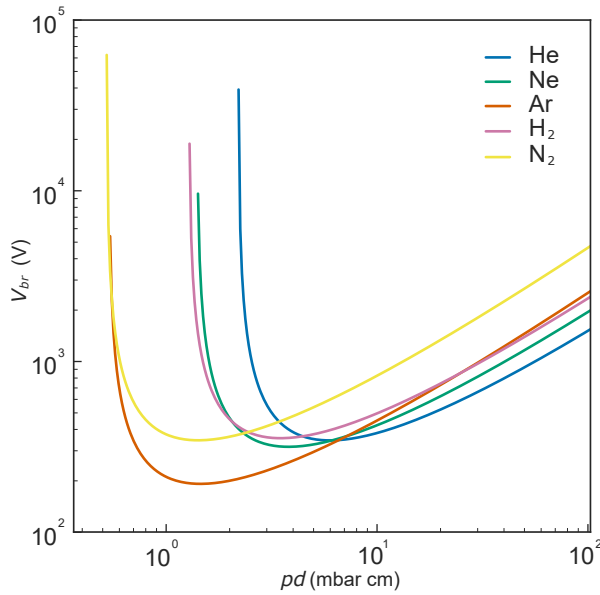


Figure 8.1: Paschen curves for several common gas types. Parameters used for calculation from [138].

discharges can be initiated at different positions in the device, causing the actual discharge curve of the system to deviate from the ideal behaviour. Therefore, discharge tests were performed with the SRFQ in order to map out the working regime of the device. These tests were performed by varying the pressure in the gas-jet chamber and recording the 1 MHz RF voltage at which a discharge was observed. DC dragging voltages are only of the order of a few volts per RFQ segment and are therefore not significant with respect to the RF voltages applied for ion confinement. At first, these tests were performed by visually observing a typical low-pressure glow discharge, such as the one shown in Figure 8.2. The results of these tests are shown as black squares in Figure 8.3.

However, the possible existence of microscopic sparks acting as sources of ions and free electrons, unobservable by the naked eye, would further reduce the working regime. To verify this, a more sensitive observation method was employed, by connecting the quadrupole rods of the DPRFQ to a Keithley 6485 picoammeter. By recording the RF voltage, applied to the SRFQ, at which a sudden jump in the measured current of the picoammeter, the discharge curve was mapped. The results of these tests are shown as red triangles labelled ‘Argon pA 1’, referring to the use of the picoammeter.

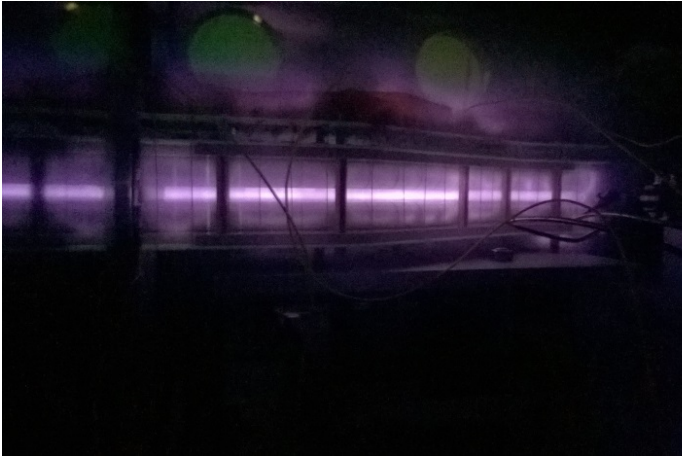


Figure 8.2: Example of an electrical discharge induced in the SRFQ.

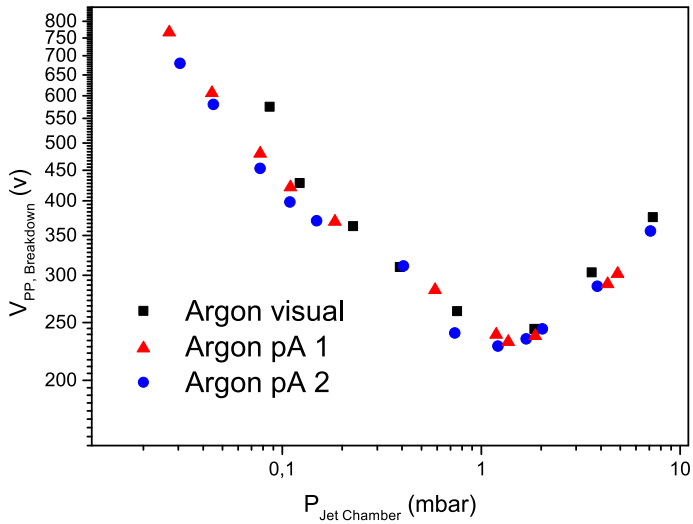


Figure 8.3: Breakdown voltages as a function of buffer-gas pressure in the SRFQ.

High-power UV lasers with energies surpassing the work function of a certain metal with which they collide could be another source of free charges that might act as nucleation for sparks. In the laser-ionization modes where at least

one of the lasers is aligned collinearly with the gas jet, a laser beam passes closely to the RFQ structure and hits the nozzle. In order to check the influence of this effect, the discharge measurements were repeated with lasers in the collinear mode. The results are shown in Figure 8.3 as blue circles labelled ‘Argon pA 2’. While the laser-induced charges do seem to have an effect on the breakdown voltages, the influence is rather small. When operating the SRFQ at different pressures, these curves have to be taken into account as upper limits to the applied RF voltage. According to the Mathieu stability parameter, q (Equations. 6.6-6.7), this is especially important when working with singly-charged, higher-mass ions. For every case in future experiments, the possibilities will have to be evaluated separately, taking into accounts the pressure in the gas-jet chamber dictated by the Mach number and stagnation pressure (Equation 5.1), as well as the ion’s mass.

8.2 Transport tests

Copper ions were used as the test case for measuring the total transport efficiency of the RFQ’s. The total efficiency is extracted by comparing the ion current measured at the beginning of the SRFQ with the ion current measured on the Faraday cup, which is positioned in the beam diagnostics chamber, as shown in Figure 8.4.

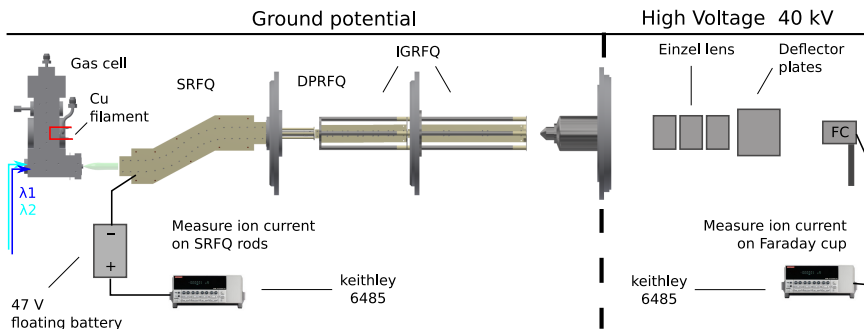


Figure 8.4: Transport measurement method.

Copper atoms were produced by resistively heating a copper filament positioned inside the gas cell, using a ≈ 30 A stabilized current. The gas cell was kept at 300 mbar with a purified (getter-based MonoTorr phase II 3000 purifier) argon gas flow kept fixed at a 1.7 slm flow by a Brooks gf series flow controller. The two Nd:YAG-pumped dye lasers were tuned to resonantly excite and ionize

copper making use of the laser ionization scheme shown in Figure 8.5. The lasers were operated at a 1 kHz repetition rate, with laser powers in the order of $\approx 50 \mu\text{J}$ and $\approx 200 \mu\text{J}$ for the first and second step, respectively. This power is high enough to saturate the first step, but this is not the case for the much weaker ionizing step (rel. intensity factor 5×10^{-4} [84]). By making use of broadband lasers of $\approx 5\text{GHz}$ bandwidth, the absolute number of ions produced was maximized as this fully covers one of the two doublet structures present in the hyperfine spectra of $^{63,65}\text{Cu}$.

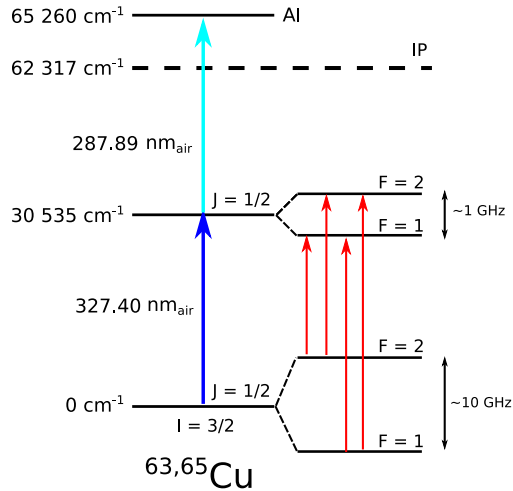


Figure 8.5: Ionization scheme of copper used for resonant ionization.

The lasers were sent into the gas cell, where they were overlapped in the ionization channel (as shown in Figure 8.4) with copper atoms seeded in the gas flow. The ions were then extracted from the gas-cell by a de Laval nozzle with a 1 mm throat diameter, designed for producing a Mach 3.5 jet. However, as the background pressure was varied over a range of 0.01 to 0.1 mbar, the supersonic jet was not well-formed (see also the operational diagram presented in Figure 8.8). This was also confirmed later by planar laser induced fluorescence measurements of this same nozzle performed by Sasha Zadornaya [134], used for visualization and characterization of the jet.

The gas cell was positioned 3 cm from the SRFQ. By connecting the negative terminal of a floating 49 V battery to the SRFQ rods, an attractive potential is created that collects the ions expelled from the gas cell. The positive terminal is connected to the signal input from a Keithley 6485 picoammeter. A detectable current is measured when it flows through the battery and the picoammeter to ground. The data was transferred by an Ethernet connection to an Advantech

serial device of which virtual COM ports can be mapped on a network computer. By blocking and unblocking the lasers entering the gas cell, the ion flow can be stopped almost instantaneously. The result of such a blocking-unblocking series of measurements on the SRFQ rods is shown in Figure 8.6. While the background signal is subject to large fluctuations in measured current, a clearly-distinguishable laser-related signal can be observed. Panel 1a of Figure 8.6 shows the raw data as measured on the SRFQ rods. In panel 1b, the same measurement is shown, with the polynomial green background shown in panel 1a is subtracted from the raw data, along with the results of a fitting with a trapezoid indicated in red.

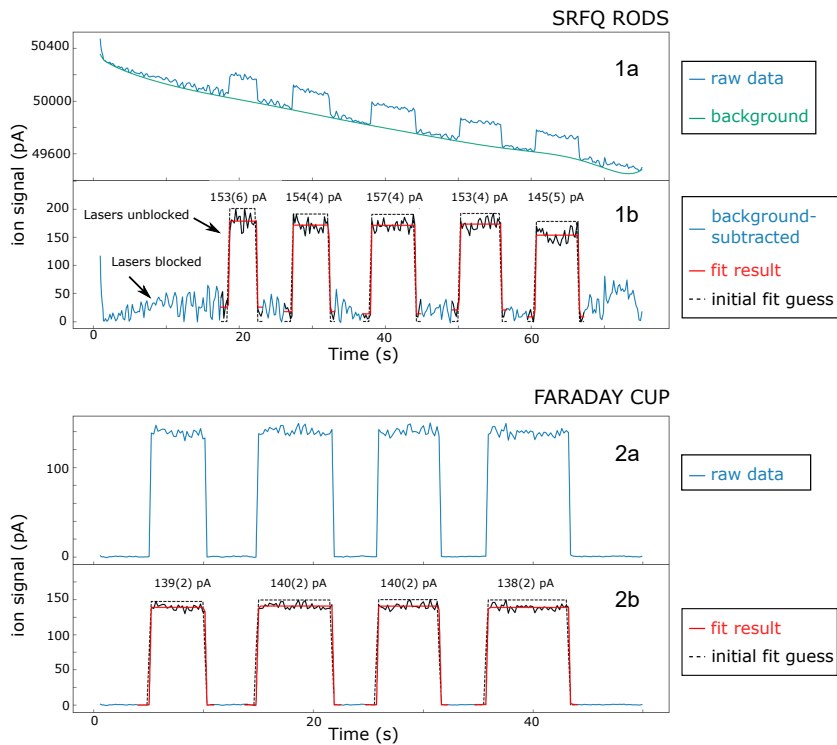


Figure 8.6: Example of a transport measurement performed with laser-ionized copper isotopes.

When these measurements on the rods of the SRFQ were taken, the battery was disconnected and the RF and DC voltage necessary for guiding copper ions were applied to all RFQ's. The ions passing through the RFQ's were then accelerated

with an electrostatic potential of 40 kV. They were focused with an einzel lens into the Faraday cup positioned in front of the mass-separating dipole magnet. This Faraday cup is connected to another Keithley 6485 picoammeter that was sent data via the network in an identical setup as discussed previously, but placed on the high voltage platform. The Faraday-cup readout is a much more stable current measurement than the battery-connection method as is clear from panels 2a and 2b of Figure 8.6, where a background is absent. After measuring on the FC, the measurement on the SRFQ rods was repeated to account for potential drifts in ion current. The ratio of the ion current measured on the FC (panel 2b) with respect to the one measured on the SRFQ rods (panel 1b) then provides the total RFQ transport efficiency. A maximal overall transport efficiency of 94(3)% was obtained.

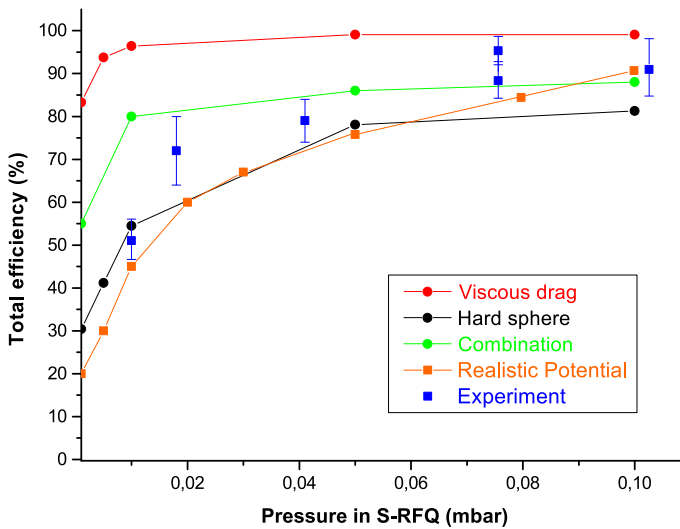


Figure 8.7: Total transmission efficiencies of $^{63,65}\text{Cu}$ ions measured at different pressures in the jet chamber compared to simulations performed with SIMION and IonCools.

The total transport efficiency was measured for different pressures ranging from 0.01 to 0.11 mbar and are compared to simulation results of both SIMION and IonCool in Figure 8.7, for which different ion-buffer gas collision models were applied. The viscous drag, hard sphere and realistic potential models were already explained in Section 6.2. The combined model, indicated by the green data points in Figure 8.7, refers to a combination of the viscous drag model applied at higher gas pressures (i.e within the jet chamber of the frontend) and the hard-sphere model applied at lower pressures (i.e. in chambers DP1 and

DP2). While the overall curves for different models agree with the down-sloping trend in efficiency for reducing pressures, the viscous drag models seems to overestimate performance, particularly at lower pressures. This is because of the ‘overdamping’ effect as noted in [126] and the lack of RF-heating effects in the model. The down-sloping trend is an effect of the reduced time the ions spend in the SRFQ for lower pressures (see also Section 8.4). Due to the smaller transit time the ions spend in the cooling region, the overall cooling effect is reduced and ions are more easily lost. The differential-pumping transitions from the SRFQ to the DPRFQ and from chambers DP1 to DP2 are the most crucial in this respect. Overall, the simulation results of the realistic potential and of the hard-sphere model seem to reproduce experimental transmission efficiency well.

Combining the transport measurements shown in Figure 8.7 with the jet-chamber pressure dictated by the Mach number and the stagnation pressure, it is possible to create an operational diagram for copper in which the setup can be operated. This diagram is shown in Figure 8.8.

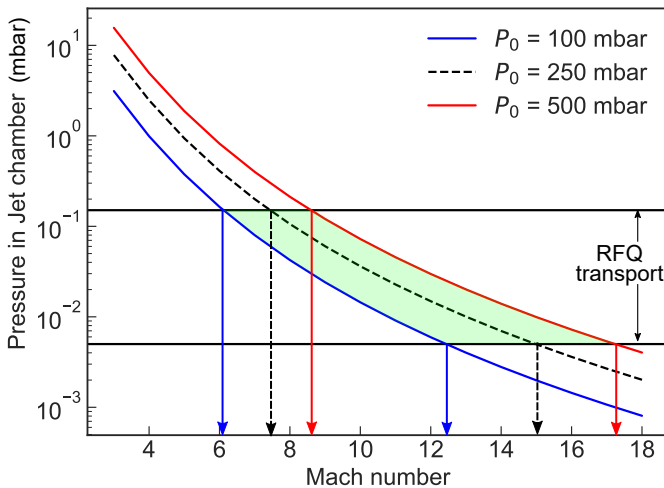


Figure 8.8: Operational diagram of the setup. The RFQ-transport limitations (horizontal solid black lines) are combined with the optimal jet-formation pressures in the jet chamber for different Mach numbers and stagnation pressures P_0 . The optimal operational regime is shown in light green. Downwards-pointing arrows delineate the Mach-number range that can be probed for $P_0 = 100$, 250 and 500 mbar shown in solid blue, dashed black and solid red lines respectively.

8.3 Longitudinal E-spread

A measure for the longitudinal energy spread of the ions was obtained by applying a blocking voltage on the differential pumping barrier separating DP1 and DP2. When ions do not have enough energy to surpass the potential barrier that is formed, the observed transport efficiency reduces. From the derivative of the S-shaped blocking curve, the longitudinal energy spread of the ion beam is measured. The energy spreads were measured for different pressures inside the jet chamber. An example of such a measured curve is shown at the bottom of Figure 8.9.

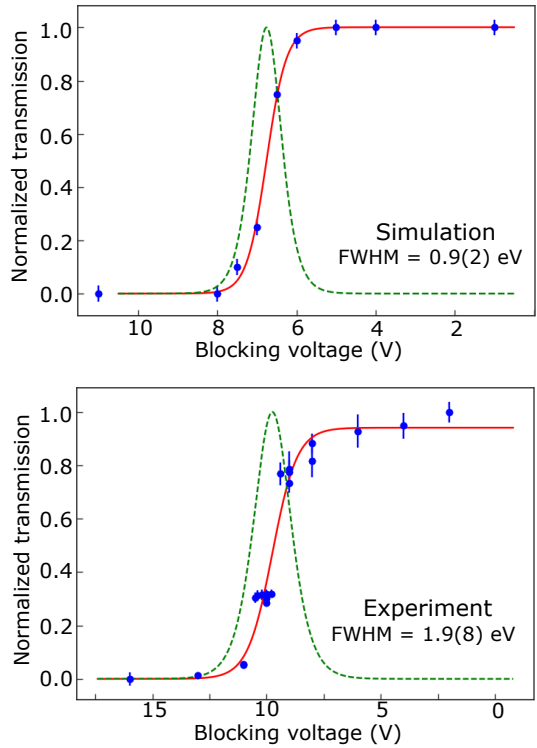


Figure 8.9: Bottom: measurement of the energy spread by varying the transmission as a function of blocking voltage applied at the second pumping barrier. The blocking voltage is measured with respect to voltage applied on the first IG electrode. Top: simulation of the experiment using IonCool.

The energy spread of the measured ions is similar to the simulated one, shown at the top of Figure 8.9. The discrepancy between the absolute blocking voltages could be explained by slightly different gradient voltage settings that were applied during the experiment with respect to the simulation. Due to the inaccuracy of the home-made voltage supplies that were used in these tests, the errors on the energy spread are large. In Figure 8.10, the energy-spread results as a function of pressure are compared with those of simulations performed using the realistic potential model with IonCool. The trend that is observed in simulations, where the energy spread decreases with increasing pressure, is not reproduced in experiment.

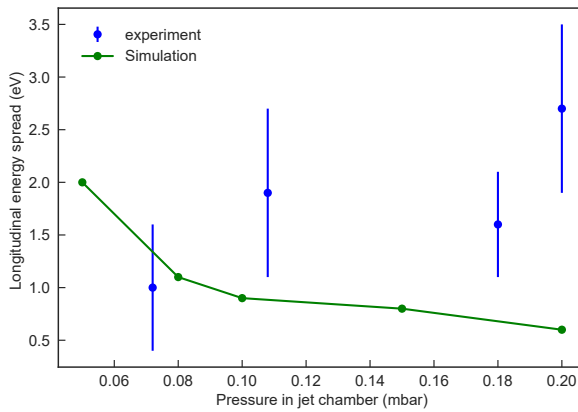


Figure 8.10: Measurement of the energy spread for different pressures.

8.4 Time profile measurements

Time profiles of mass-separated ions arriving on an MCP detector ($\varnothing 25$ mm Gids GmbH) positioned at the focal plane of the dipole-magnet mass separator were taken with the experimental setup shown in Figure 8.11. The MCP consists of a two-plate chevron type setup placed in front of a phosphor plate (type P43). The phosphor can both be used for imaging and as an anode for ion-current detection. The latter was used for obtaining the time profiles of laser-ionized $^{63,65}\text{Cu}$ and Cu-compounds that were formed either in the gas-cell or in the RFQ's. The MCP was biased to single-ion detection mode at -2100 V while the phosphor plate was kept at ground potential. The signal from the phosphor

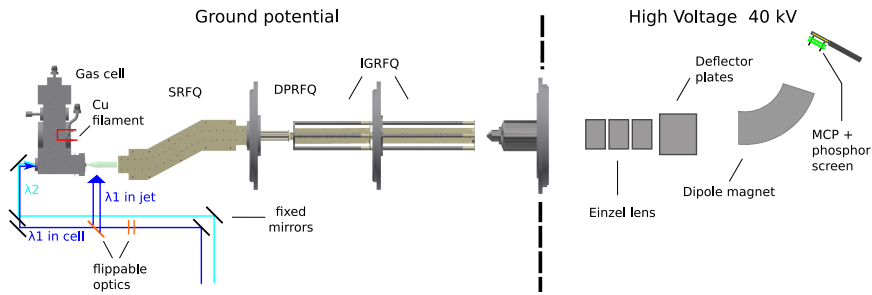


Figure 8.11: Time profile measurement method.

plate was sent through an Ortec fast preamp VT 120 before it was passed to a timing filter amplifier and multichannel analyser (MCA). The start signal of the time profiles was given by the laser-pulse trigger and time profiles for both in-gas-jet and in-gas-cell ionization were taken. When the in-gas-cell method was employed, both lasers were sent through the gas cell from a window at the back of the cell before they exited via the nozzle. In this way, both in-gas-jet and in-gas-cell components are present in the time profiles, as both are contained within the ionization volume. Before entering the cell, the lasers passed a 1 mm diameter pinhole thereby overlapping only with the center of the 10 mm diameter final gas-cell channel. This reduces the in-cell ionization component that would otherwise be much larger than the in-jet component with such a setup. An example of time profiles for ^{65}Cu obtained with the latter method and with in-gas-jet ionization is shown in Figure 8.12, when the magnet was tuned to $A/Q = 65$. For in-gas-jet ionization, the time profile shows a single peak arriving at the MCP detector in the order of 1.5 ms after the ions were created. The in-gas-cell time profile consists of two components which can be attributed to ionization in the jet or close to the nozzle (fast component) and to a slow component of ions created in the cell and extracted by the much slower gas flow (slow component).

Ions created using the in-gas-jet method spend most of their time within the SRFQ, as the pressure here is the highest. Due to this, the influence of the DC voltage gradient applied between the first and last electrode of the SRFQ upon the time profiles of the extracted ions was investigated. In Figure 8.13, the voltage at the back of the SRFQ was kept at -32 V, while the labels indicate what the voltage was at the start of the SRFQ. During this test, the pressure in the jet-chamber was kept at 2.2×10^{-2} mbar. As can be seen from this figure, it is possible to shift the arrival time from 0.65(4) ms (FWHM 0.12(4) ms) at a 32 V SRFQ gradient to a 4.8(2) ms (FWHM 2.9(2) ms) at 0 V gradient, while keeping the total integral of all curves almost constant.

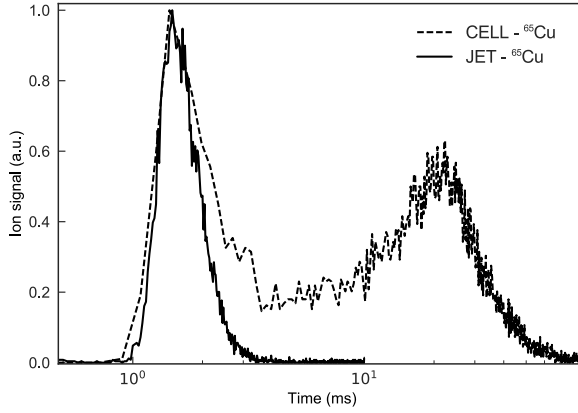


Figure 8.12: Normalized (peak value = 1) time profiles of ions created using in-gas-cell ionization (setup explained in text) and in-gas-jet ionization.

The only (expected) exception appears when the voltage gradient was reversed by applying -35V at the SRFQ entrance. The blue curve indicating this results disappears in the background, as the transport efficiency drops to zero.

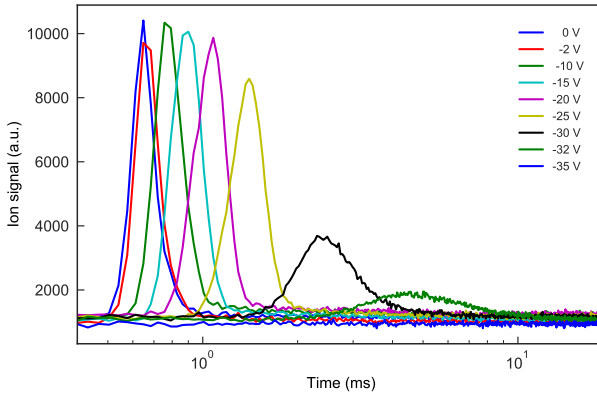


Figure 8.13: Time profiles obtained when changing the SRFQ voltage gradient.

These tests were repeated for four different pressure at different voltage gradients. The results of these measurements are shown in Figure 8.14. While in experiment,

the pressure was only varied over a small range ($1 - 4 \times 10^{-2}$ mbar), it has a large impact on the observed time profiles. For the 2 V gradient, the arrival time ranges from 2.4(2) to 6.0(5) ms, while for the 32 V gradient they range from 0.64(8) to 1.3(2) ms.

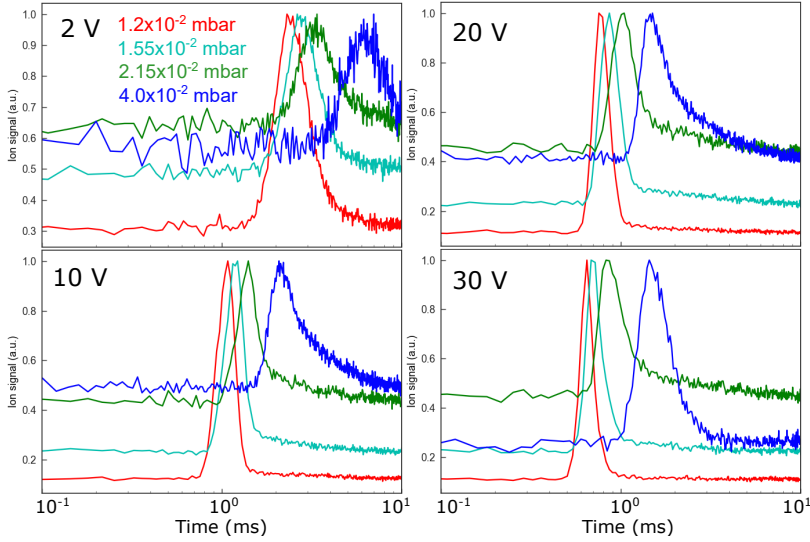


Figure 8.14: Transport times of ^{63}Cu at different pressures and voltage gradients across the SRFQ. Voltage gradients are indicated in the top left corner of each panel. In order to provide a fair comparison of the time profiles, since the filament current and laser power varied for the different tests, the maximum of their value of their time profile was all normalized to 1.

Time profiles are summarized and compared to IonCool simulations making use of the hard sphere (full line) and realistic potential (dashed line) in Figure 8.15. From this plot, it is seen that the realistic potential seems to provide a much better agreement with experimental data.

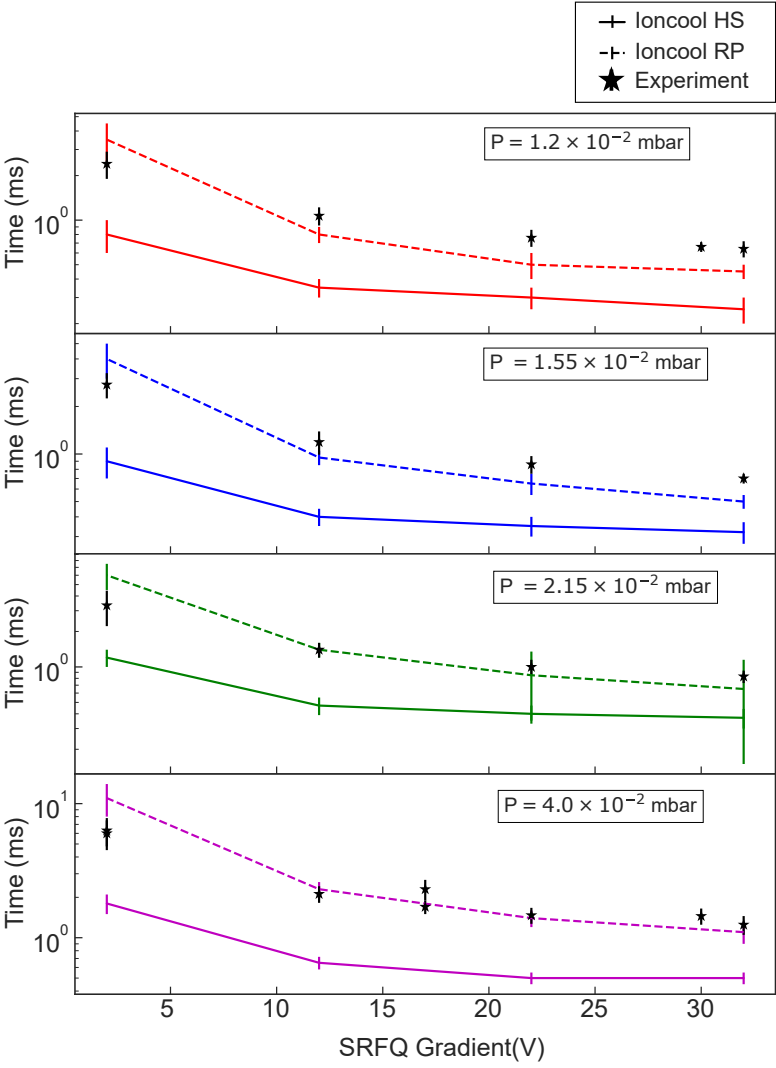


Figure 8.15: Comparison of experimental and calculated transport times using IonCool, with either the hard sphere (HS) or realistic potential (RP) models.

8.5 Emittance measurements

The emittance is a measure of the quality of an ion beam and is related to its volume in coordinate and momentum phase space. This value is most often quoted as the surface of an ellipse covering 95% of the beam phase space volume plotted as a function of position coordinates i perpendicular to the beam direction and momentum coordinates $i' = v_i/v_z$ where $i = x$ or y when the beam moves in the z direction

Transversal emittance measurements were performed to further characterize the ion-beam quality coming from the in-gas laser ion source and RFQ's. The tests were performed using a method detailed in [140–142]. This type of measurement only makes use of an electrostatic focussing element, which in this case was an einzel lens, and a beam imaging technique. The setup used for measuring the emittance is shown in Figure 8.16. By scanning the einzel lens voltage and recording the beam-spot width as a function of the lens voltage, and combining this with results from simulations, it is possible to extract the transversal emittance.

An ion beam can be characterized by its Σ -matrix

$$\Sigma = \begin{pmatrix} \sigma_x^2 & \sigma_{xx'} \\ \sigma_{xx'} & \sigma_{x'}^2 \end{pmatrix} = \begin{pmatrix} \sigma_{11} & \sigma_{12} \\ \sigma_{21} & \sigma_{22} \end{pmatrix} \quad (8.1)$$

where $\sigma_x^2(\sigma_{xx'})$ is the variance (covariance) of the coordinate distributions. The rms-emittance of an ion beam is determined as $\varepsilon_{\text{rms}} = \sqrt{\det(\Sigma)}$. The combined action of electrostatic elements of a beam line on an ion beam moving from position z_0 to z_1 can be calculated as matrix multiplication with a transfer matrix R

$$R = \begin{pmatrix} R_{11} & R_{12} \\ R_{21} & R_{22} \end{pmatrix} \quad (8.2)$$

The description of the ion beam at position 1, Σ^1 , is then related to R and Σ^0 as

$$\Sigma^1 = R\Sigma^0 R^T. \quad (8.3)$$

The expression for the σ_{11}^1 that can be measured at the MCP position when this matrix product is written out is given as

$$\sigma_{11}^1 = R_{11}^2 \sigma_{11}^0 + 2R_{11}R_{12} \sigma_{12}^0 + R_{12}^2 \sigma_{22}^0 \quad (8.4)$$

This means that, as the beam width σ_x^2 is measured at position 1 (MCP position) for different einzel lens voltages, this relation can be used to calculate the matrix Σ^0 , by solving a set of equations, assuming the matrix elements of R are known. In order to obtain the matrix elements of R , IonCool simulations were performed for the IGLIS setup with simple ion starting conditions. Ions started either on axis, with an angle α or started off-axis without a velocity component in the transverse direction ($\alpha=0$). These simulations yield the matrix elements of R as linear functions of the einzel lens voltage.

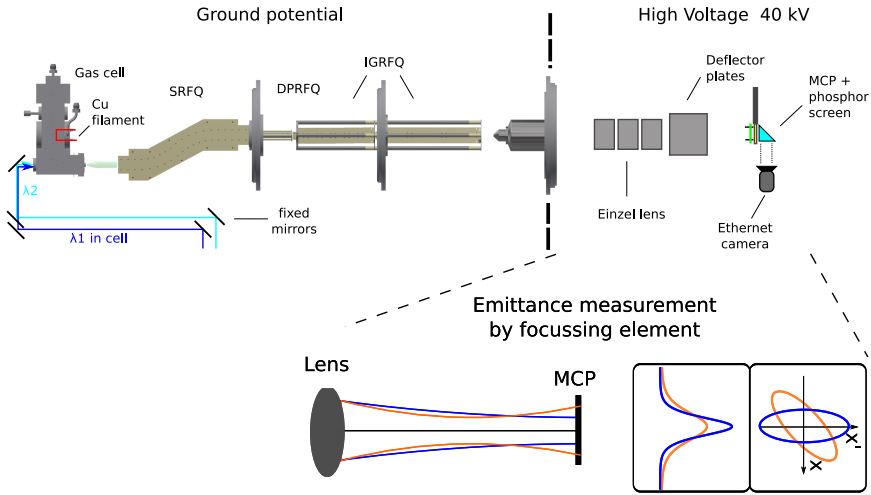


Figure 8.16: Emittance measurement setup.

The beam spot is recorded with an IDS Ethernet camera pointed at a prism located at the back of the phosphor screen on which the beam spot is projected. The picture is then analysed by projecting the image intensity on the x and y axes and fitted with a Gaussian curve as shown at the top of Figure 8.17. The result of varying the einzel lens voltage on the beam spot size is given on the bottom of Figure 8.17.

A quadratic fit, from which the parameters were used for calculating the final y-emittance is shown in red. The resulting emittance was found in this case to be $\varepsilon_y = 12(2)\pi$ mm mrad and $\varepsilon_x = 8(3)\pi$ mm mrad. These values agree in order of magnitude with the values obtained in simulations as shown in Figure 8.18.

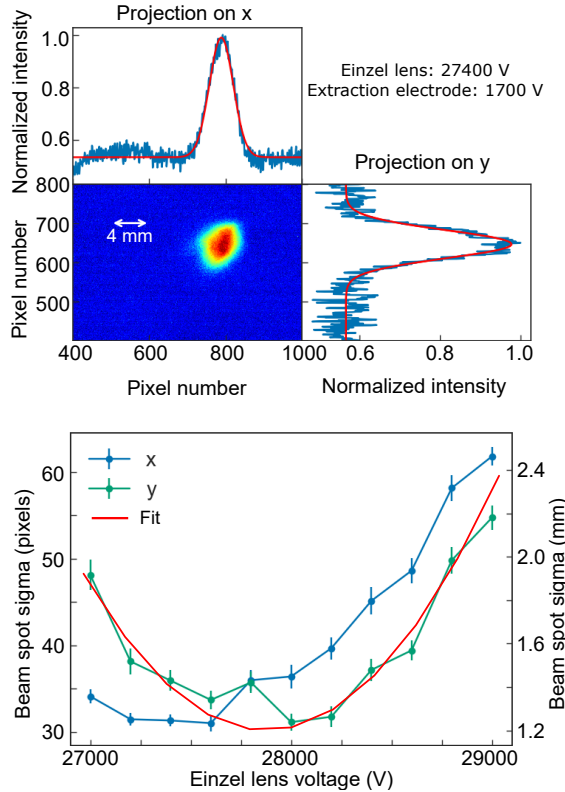


Figure 8.17: Top: Example of a beam spot recorded at the MCP positioned before the dipole magnet at an einzel lens voltage of 27400 V at a jet-chamber gauge pressure of 4×10^{-2} mbar and 32 V SRFQ gradient. Bottom: Beam spot size as a function of einzel lens voltage with fit results shown in red.

Similar emittance measurements were performed at different pressures in the gas-jet chamber to study the influence of buffer-gas cooling on the emittance. The results of these tests are shown in Figure 8.20. However, at the time at which these experiments were performed, a considerable amount of steering was present in the beam line. The only combination of extraction electrode and

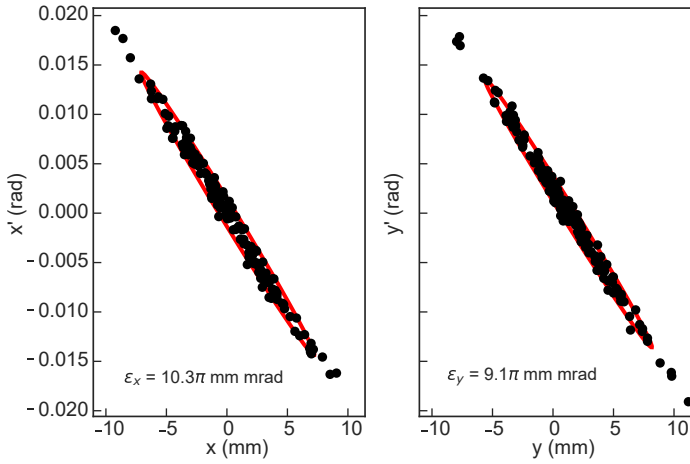


Figure 8.18: IonCool simulated emittance of ^{63}Cu at the MCP position, for the same pressure as in Figure 8.17, with a SRFQ gradient of 32 V.

einzel lens voltages for which a minimum of beam diameter could be observed at the MCP, was when the extraction electrode was reduced to 400 V and the einzel lens was varied from +26000 to +28000 V with respect to the -40 kV high-voltage offset ground potential (i.e. -14 kV to -12 kV with respect to ground). However, these voltage settings induced large beam aberrations, such as the one shown on the left-hand side of Figure 8.19. When going to higher voltage settings of the einzel lens, steering caused the beam spot to deviate from the MCP center. An example of this is shown on the right-hand side of Figure 8.19. The latter effect causes the asymmetry in the curves of Figure 8.20.

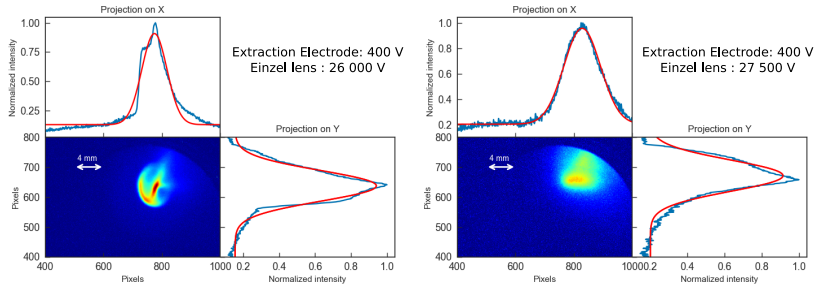


Figure 8.19: Left: Example of a beam spot suffering from strong aberrations measured at the MCP detector for extraction electrode and einzel lens voltages of 400 and 26000 V respectively. Right: Effect of steering causing the beam to only partially hit the MCP detector at higher einzel lens voltages.

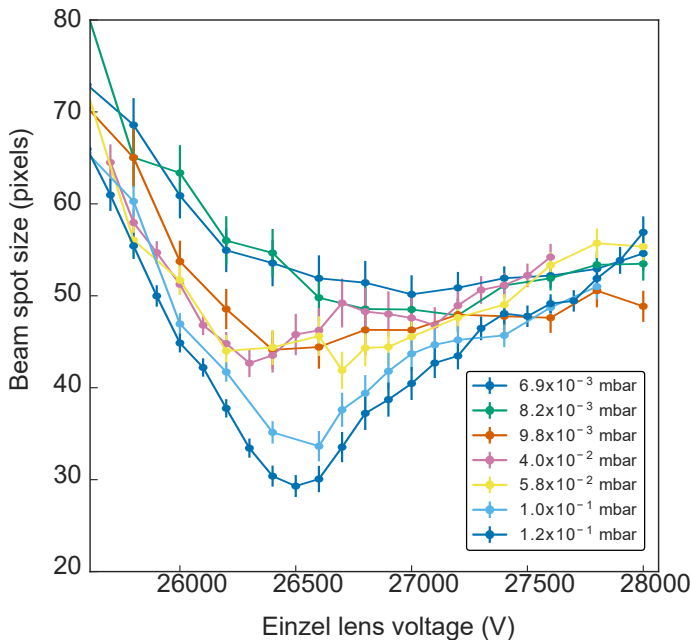


Figure 8.20: Recorded beam sizes for different pressures inside the SRFQ.

However, a clear pressure dependence is observed from these measurements, as the beam-spot size is steadily reduced for higher pressures. This agrees with

what would be expected as shown in Figure 8.21, where the emittance results of IonCool simulations are shown for different jet-chamber pressures. In order to be able to compare experimental results with these simulations, further tests should be performed without the effects of steering and beam aberration being present.

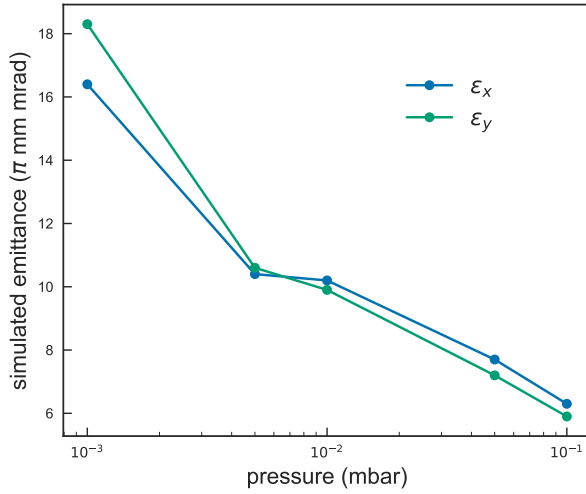


Figure 8.21: Simulated emittances for ^{63}Cu at the MCP position for different pressures with a 22 V SRFQ gradient. Simulations were performed with the realistic potential model in IonCool.

8.6 Mass scans and contamination

In the final stages of the commissioning of the laser- and jet laboratories for IGLIS studies, a series of mass scans were taken to characterize the beam composition extracted from the in-gas laser ion source and RFQ's. The spectra were recorded on a MCP detector positioned at the focal plane of the IGLIS jet laboratory beam line. A 1-mm wide slit was placed in front of the 25mm diameter MCP detector to increase the mass resolving power ($\Delta m/m$) to 250(50). A phosphor plate at the back of the MCP was used as the anode which was connected to an MCA counter module.

Ionization in a gas cell is a process in which many different processes happening inside the cell can affect the composition of the extracted ion beam. In the many collisions taking place inside the gas cell with buffer-gas or other contaminants, the atoms of interest can form molecular bonds or get lost to a dark state, which cannot be probed with the lasers used for ionization [143]. After nearly-instantaneous ionization by a short laser pulse inside the cell, the ions are extracted out of the cell. During this flow, neutralization losses due to a charge-exchange process can occur. Another possibility is that the ions bond with atoms or molecules in the gas and form adducts. A mass spectrum extracted from a gas-cell-based ion source can therefore be rich in adducts, depending on its chemistry and the presence of impurities in the gas. Because of this, it is obvious that using a high-purity gas and a well-conditioned and clean setup is of great importance. For the IGLIS in-gas-jet laser ion source, the spectrum is expected to contain much less of such adducts, as the pressure differences is of at least two orders smaller in magnitude [$O(100 \text{ mbar}) - O(\leq 1 \text{ mbar})$]. The time an ion spends inside the gas in gas-cell ionization is also an order of magnitude larger ($O(10 \text{ ms}) - O(1 \text{ ms})$): see also Section 8.4).

Mass scans that were taken using in gas-cell and in-gas-jet laser ionization are compared on the right side of Figure 8.22. For in-gas-cell ionization, both lasers were sent into the cell from the back, while for in-gas-jet ionization, laser 1 was formed into a laser sheet and was directed in a perpendicular direction into the gas jet. In the linear-scale plots 1a and 2a of Figure 8.22, four intense doublets are observed in gas-cell measurements. The lowest-mass doublet comes from $^{63,65}\text{Cu}$, while the others are related to adduct formation with either one or two water molecules or with argon. The peaks related to $\text{Cu}+2[\text{H}_2\text{O}]$ are even larger than the pure copper peaks. The peaks are coming from contaminants present in the gas, with which Cu^+ is able to form strong bonds. The most important observed adducts are shown in Table 8.1.

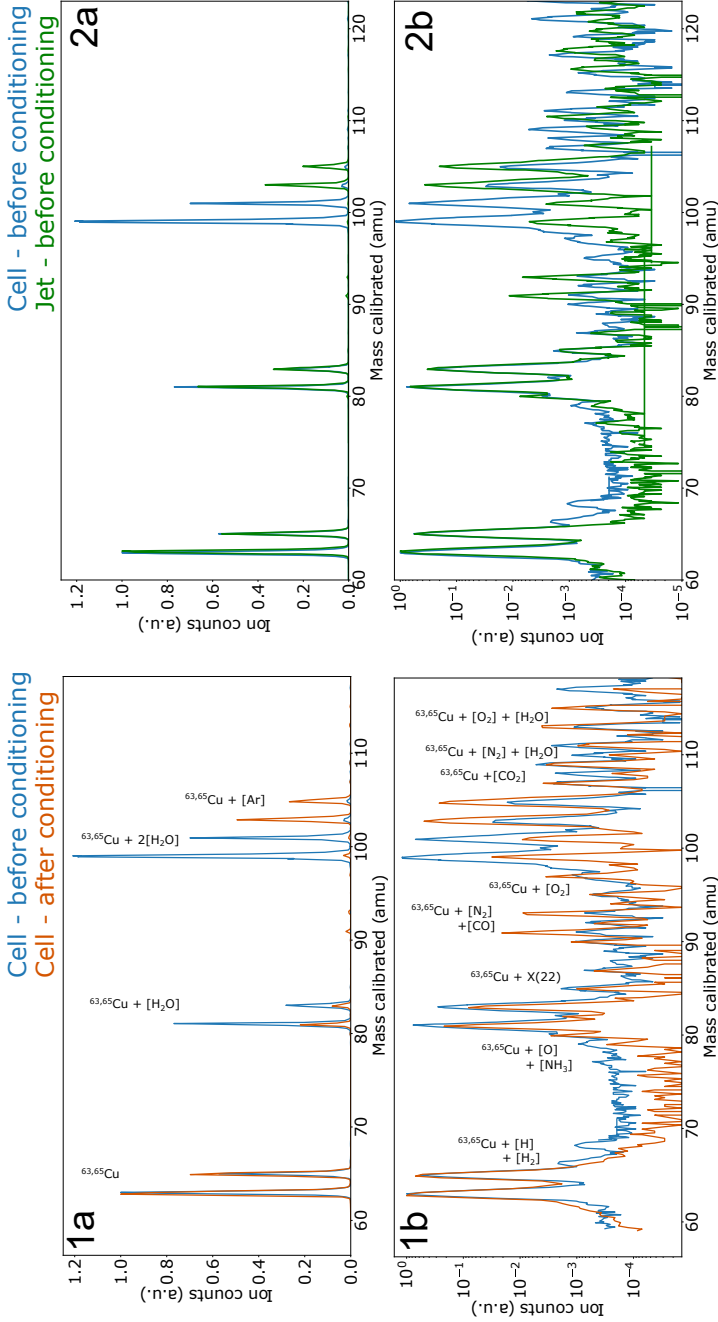


Figure 8.22: Left: Mass scans obtained with both in-gas-cell laser ionization before and after conditioning of the gas cell in linear (1a) and log scale (1b). Right: Mass scans obtained with in-gas-cell (2a) and in-gas-jet (2b) laser ionization both taken before conditioning of the cell shown in linear (2a) and log scale (2b). Labels indicate the most likely origin of the copper adducts.

Table 8.1: Summary of Cu-adducts formed with impurities present in the gas and their dissociation energy. Data from [144] and [136]

Adduct	Mass A	Dissociation energy (kJ/mol)
$^{63,65}\text{Cu}^+ - \text{H}$	64, 66	93(13)
$^{63,65}\text{Cu}^+ - \text{H}_2$	65, 67	51.9(4)
$^{63,65}\text{Cu}^+ - \text{CH}_2$	77, 79	267.3(68)
$^{63,65}\text{Cu}^+ - \text{CH}_3$	78, 80	111(7)
$^{63,65}\text{Cu}^+ - \text{O}$	79, 81	134(12)
$^{63,65}\text{Cu}^+ - \text{H}_2\text{O}$	81, 83	160.7(75)
$^{63,65}\text{Cu}^+ - \text{NH}_3$	80, 82	237(15)
$^{63,65}\text{Cu}^+ - \text{N}_2$	91, 93	89(30)
$^{63,65}\text{Cu}^+ - \text{CO}$	91, 93	149(7)
$^{63,65}\text{Cu}^+ - \text{Ar}$	103, 105	52(10)

In the spectra of Figure 8.22, all peak intensities were normalized to ^{63}Cu . It should be stated that, while peak intensities can be compared relative to one another for spectra taken under similar conditions, the relative peak heights within a single spectrum do not necessarily correspond to the actual intensity present in the beam. The reason is that all masses might not be transmitted through the RFQ’s with the same transport efficiency. If one wished to study the relative intensity of all adducts extracted from the gas cell, the RFQ systems should be optimized for all different masses for which the produced amount would be studied. However, the change in intensity from one condition to the other, as is presented here in these cases, can be compared (e.g. for in-cell and in-jet ionization in panel 2a/b).

The most striking difference between the two spectra shown in panel 2b of Figure 8.22, is seen in peak of $\text{Cu}+2[\text{H}_2\text{O}]$. This component is reduced by a factor 240, while the $\text{Cu}+[\text{H}_2\text{O}]$ peaks remain present. The peaks related to Cu-Ar adducts seems to have increased. The reason for this is not immediately clear.

In panels 1a and 1b of Figure 8.22, the blue curve is the same as the one shown in panels 2a and 2b. It is now compared to a spectrum that was taken using the in-gas-cell ionization method, after the setup was conditioned for two consecutive days. During this conditioning the gas cell and gas transfer lines were baked out by heating them to $\approx 100^\circ\text{C}$. The effect seems to be remarkably similar to the in-gas-jet spectrum that was just discussed. While the $\text{Cu}+2[\text{H}_2\text{O}]$ peaks show a steep decrease before and after conditioning, $\text{Cu}+[\text{H}_2\text{O}]$ is still present and the intensity of Cu-Ar adducts increased. However, the relative

difference before and after conditioning is slightly larger here for $\text{Cu} + [\text{H}_2\text{O}]$. The unconditioned $\text{Cu} + [\text{H}_2\text{O}]$ peak was only 1.15 times larger than the peak for in-gas-jet ionization, while this has now changed to a factor 3.45.

The reason for this might be that the reaction rate between copper and H_2O is high enough to form adducts even in the low-pressure regime of the RFQ's, where many collisions occur with the buffer gas. On the other hand, $\text{Cu} + 2[\text{H}_2\text{O}]$ takes a longer time to form and grow in intensity, as shown by the time profiles obtained for different adducts in Figure 8.23. This can only happen if the concentration of H_2O is high enough.

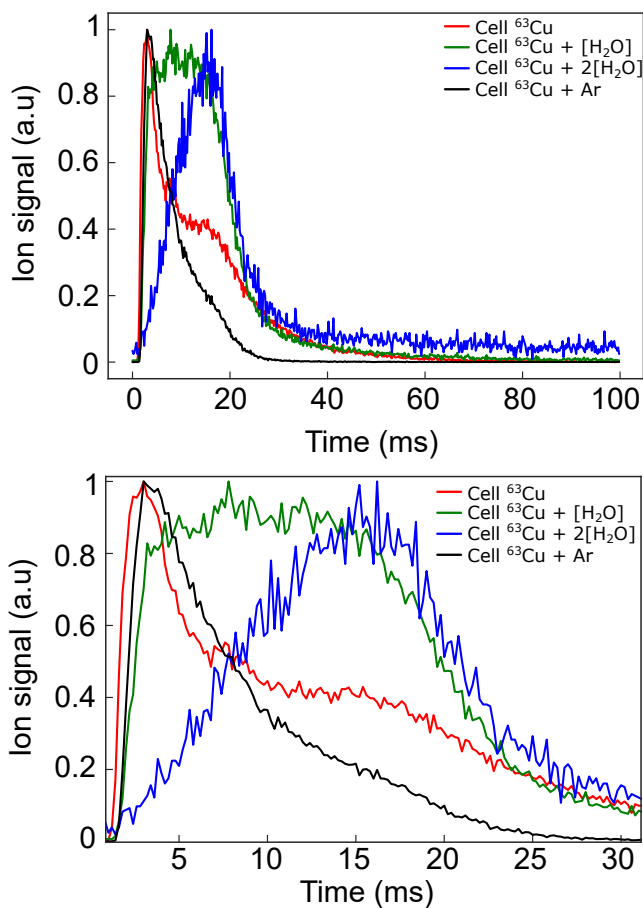


Figure 8.23: Top: Time profiles for different ^{63}Cu adducts produced via in-gas-cell ionization measured after mass separation. Bottom: Zoomed-in version of the same data as presented in the top plot.

The reason for this may be that in order to form a measurable $\text{Cu} + 2[\text{H}_2\text{O}]$ contribution via two-body collisional processes, the concentration of both $\text{Cu} + [\text{H}_2\text{O}]$ itself and of H_2O has to be high enough. This former concentration is related the involved time scale for $\text{Cu} + [\text{H}_2\text{O}]$ adduct-formation that depends both on the reaction rate constants and on the concentration of the Cu^+ and H_2O reactants [143]. Here, a strong difference in the time profiles of observed adduct formation was observed, related to the impurity concentration.

From these tests, it's easily concluded that conditioning the gas-cell and keeping the impurity-level as low as possible is important. In the future, it would be interesting to test several molecular-dissociation methods to purify the beam content. This could be tested using either laser- or electric-field-induced dissociation.

Chapter 9

Conclusions and outlook

9.1 Conclusions

This thesis presents the results of both an in-source laser-ionization spectroscopy study of exotic mercury isotopes and the design, simulation and commissioning of RFQ ion guides for the development of the in-gas-laser ionization and spectroscopy (IGLIS) technique.

9.1.1 Laser spectroscopy of mercury

The aim of the laser-spectroscopy experiment was to study the ground-state properties across a broad range of mercury isotopes through the measurement of their isotope shifts and hyperfine structures. More specifically, the goal was to observe the evolution of the previously-discovered shape staggering for nuclei more exotic than the so-far lightest studied isotope, ^{181}Hg [5]. In the neutron-rich region, the aim was to extend the knowledge of the charge radii across the closed neutron shell to verify the existence of a kink in the linear charge-radii trend at $N = 126$, as is observed in several elements in the same region with $Z > 82$ [28].

The experiment was performed at the ISOLDE facility in CERN, combining the ISOL technique with in-source laser spectroscopy. Different setups were used to measure the intensity of resonantly-ionized isotopes as a function of laser wavelength. The Windmill detection setup used the detection of α particles and γ rays resulting from the radioactive decay of the neutron-deficient nuclei.

ISOLTRAP's MR-ToF-MS device measured the ion's time-of-flight to produce quasi-background-free hyperfine spectra. The combined use of both techniques made it possible to measure a mass range from ^{177}Hg to ^{208}Hg , spanning production rates of 0.1 ions/s to several pA of beam current.

By measuring the isotope shift and hyperfine spectra, the charge-radii differences, magnetic dipole and electric quadrupole moments of these isotopes were obtained. The results for the neutron-deficient nuclei establishes the shape-staggering endpoint of mercury. It was shown from the charge-radii and deformation that the famous odd-even staggering is a localized phenomenon between $180 < A < 186$ and that mercury isotopes with $A \leq 180$ return towards sphericity.

On the neutron-rich side, a kink in the charge-radius trend at $N = 126$ was observed for the first time in an element below the closed $Z = 82$ proton shell. The experimental findings for the neutron-deficient nuclei were compared to state-of-the-art DFT and MCSM calculations. The DFT calculations offer an interpretation of the shape staggering as an emergence of multiple near-degenerate PES minima of different deformations. A qualitative agreement could be reached by tuning the parameters of either the pairing energy in a UNEDF1^{SO} functional or by changing the surface tension of SLy5x functionals. However, a quantitative reproduction is not possible due to the sensitivity of the charge radii to the functional's parameters and the exact location of the staggering is shifted by two mass units with respect to the experiment.

Large-scale MCSM calculations pointed to a strong monopole interaction between the proton $\pi h_{9/2}$ and neutron $\nu i_{13/2}$ orbitals as the reason for the large deformation in the odd-mass mercury isotopes $^{181,183,185}\text{Hg}$. Large differences in nucleon-occupation numbers of these orbitals were obtained for the odd-even staggering nuclei. The MCSM calculations were able to reproduce both qualitatively and quantitatively the experimental charge-radii, dipole- and quadrupole-moments. However, the energetic ordering and location of all the calculated energy levels are not in perfect agreement with the experimental findings. One reason for this may be the use of a limited number of basis vectors within the calculations. These calculations provide a quantitative and microscopic insight into the system, pointing to the specific nucleon orbitals responsible for the staggering phenomenon. The mechanism also links to nuclear structure phenomena in other regions of the nuclear chart, such as the quantum-phase transition in zirconium or shape coexistence in ^{68}Ni .

9.1.2 RFQ ion guides

In-gas-jet laser ionization spectroscopy is a promising technique which aims to overcome some of the limitations hindering laser spectroscopy in unexplored regions of the nuclear chart, such as the heavy- or refractory element regions. It has recently been shown that it is possible to perform in-gas-jet laser spectroscopy with an improved resolution of more than one order of magnitude, with respect to in-gas-cell spectroscopy, without losing efficiency [13]. The Heavy Element Laser Ionization and Spectroscopy (HELIOS) project at KU Leuven aims to develop this technique, which is fast, selective and sensitive, in order to couple it to the next-generation radioactive ion beam facilities using in-flight production methods, such as SPIRAL2 at GANIL. This would allow high-resolution (≈ 100 MHz) laser spectroscopy to be performed in the heavy-element region, where production yields are extremely low.

For the development of the IGLIS technique, a new laser and mass-separator laboratory at KU Leuven has been built. This thesis presents the design, simulation and commissioning of the RFQ ion guides which capture ions produced using the IGLIS technique. The RFQ's need to efficiently capture the ions, and transport them from their area of production to a high vacuum region, passing two differential pumping barriers, with a minimal amount of ion losses while providing a good beam quality. The final RFQ's consist of three separate RFQ structures that were designed for this purpose. Their properties were simulated using different software packages. Finally, they were constructed and commissioned.

In the commissioning tests, measurements of the transport efficiency, longitudinal energy spread, ion-transmission time profiles and beam emittance were performed. Voltage and pressure regimes at which the RFQ's could operate were mapped out by measuring the Paschen curve. During the best operation, a transport efficiency of 94(3)% and an energy spread of 1.0(5) eV could be measured. The emittance was found to be of the order of 9π mm mrad for a 2 keV beam energy. The test results are in agreement with the simulated properties. The operational diagram in which the RFQ's can be optimally used for gas-jet spectroscopy, is limited by its transport efficiency related to gas-jet-imposed pressure limits, as well as breakdown-imposed voltage limits. The RFQ's are currently installed in Leuven as part of the IGLIS laboratory.

9.2 Outlook

As was pointed out in the first part of this thesis, the ground-state shape staggering in the mercury isotopes is a phenomenon that is restricted to the neutron mid-shell region. Similar ground-state shape transitions to those in mercury have been observed in other elements in the neutron-deficient lead region, such as in gold [145] and iridium [146]. Within recent experimental campaigns performed at ISOLDE, we studied gold and bismuth isotopes with neutron numbers, down to $N = 97$ and $N = 104$ respectively. Analyses of these experiments are ongoing, but the preliminary results are intriguing. For gold, the isotope shifts indicate a return towards less-deformed nuclear ground states for the more neutron-deficient isotopes, while in bismuth, a shape-staggering pattern similar to mercury was observed around $N = 105$.

It would be interesting to delineate these mid-shell shape-transition regions for both the lighter elements such as iridium, osmium, rhenium, tungsten and tantalum, as well as for the heavier elements such as bismuth, polonium and astatine. However, these lighter elements are refractory metals, and their production using the ISOL technique is notoriously difficult. For the heavier elements, production yields, life times and isobaric contamination are the main experimental hindering factors. A future upgrade of the ISOLDE facility to accept 2-GeV proton beams could be of help here by increasing ion-beam yields up to a factor of 6 [147, 148]. As mentioned in **Paper II**, the shape-staggering reproduction is very sensitive to several tunable nuclear-theory parameters. As such, theory could benefit from a large dataset in this region of the nuclear chart in order to benchmark new effective interactions.

Making use of experimental techniques other than laser spectroscopy would provide complementary measurements of the nuclear structure in the region of isotopes around mercury. Recently, Coulomb-excitation studies have been performed for the even-mass mercury isotopes $^{182-188}\text{Hg}$ in the safe Coulex regime [149]. Extending these measurements to the odd-mass isotopes would provide independent measurement of deformation of the ground and excited states. Transfer-reaction experiments using beams of exotic mercury in inverse kinematics could be performed at for instance the HIE-ISOLDE facility using the recently-installed ISOLDE Solenoid Spectrometer [150]. Such measurements probe directly the single-particle nature of the populated states [151]. In the future, electron-scattering experiments in storage rings such as the SCRIT-experiment in RIKEN [152], could be used to study charge-density distributions of exotic nuclei from which absolute charge radii can be extracted.

Improving the resolution of current laser-spectroscopy measurements would increase the sensitivity to the quadrupole moments of the measured isotopes, which is generally low in in-source spectroscopy. This can be achieved using either conventional collinear laser spectroscopy, or one of several techniques under development such as PI-LIST [14], Doppler-free two-photon spectroscopy [153, 154] or MIRACLS [15]. Currently, the sensitivity of these experiments is not high enough to reach e.g. ^{177}Hg . However, technical developments are continuously ongoing.

The second part of this work already hinted towards yet another outlook for laser-spectroscopy studies, as the RFQ's presented in this thesis form part of a dedicated IGLIS setup with the goal of studying unexplored regions of the nuclear chart. The IGLIS technique can be used in current facilities producing exotic ion beams via in-flight-separation [155] or in new facilities such as SPIRAL2 [156] that are starting up. Here, the in-gas-jet laser ionization technique could be used for the production and study of exotic isotopes in the heavy elements, potentially up to rutherfordium [13] or in the refractory element region. This would further the reach of laser spectroscopy across the nuclear chart, providing new access to key regions of interest in the nuclear landscape, using a near-model independent probe. Key studies in these regions could include ground-state shape evolutions in the refractory elements, the evolution of magic numbers in exotic nuclei, the study of exotic $N = Z$ -nuclei with amongst others ^{100}Sn and heavy and superheavy nuclei, including nobelium isotopes and above.

Bibliography

- [1] B. A. Marsh, T. D. Goodacre⁺, **S. Sels**⁺, Y. Tsunoda, B. Andel, A. N. Andreyev, N. A. Althubiti, D. Atanasov, A. E. Barzakh, J. Billowes, K. Blaum, T. E. Cocolios, J. G. Cubiss, J. Dobaczewski, G. J. Farooq-Smith, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, L. P. Gaffney, L. Ghys, M. Huyse, S. Kreim, D. Lunney, K. M. Lynch, V. Manea, Y. M. Palenzuela, P. L. Molkanov, T. Otsuka, A. Pastore, M. Rosenbusch, R. E. Rossel, S. Rothe, L. Schweikhard, M. D. Seliverstov, P. Spagnoletti, C. V. Beveren, P. V. Duppen, M. Veinhard, E. Verstraelen, A. Welker, K. Wendt, F. Wienholtz, R. N. Wolf, A. Zadvornaya, and K. Zuber, “Characterization of the shape-staggering effect in mercury nuclei,” *Accepted in Nature Physics*, 2018, ⁺ these authors contributed equally to this work.
- [2] **S. Sels**, T. D. Goodacre, B. A. Marsh, Y. Tsunoda, B. Andel, A. N. Andreyev, N. A. Althubiti, D. Atanasov, A. E. Barzakh, J. Billowes, K. Blaum, T. E. Cocolios, J. G. Cubiss, J. Dobaczewski, G. J. Farooq-Smith, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, L. P. Gaffney, L. Ghys, M. Huyse, S. Kreim, D. Lunney, K. M. Lynch, V. Manea, Y. M. Palenzuela, P. L. Molkanov, T. Otsuka, A. Pastore, M. Rosenbusch, R. E. Rossel, S. Rothe, L. Schweikhard, M. D. Seliverstov, P. Spagnoletti, C. V. Beveren, P. V. Duppen, M. Veinhard, E. Verstraelen, A. Welker, K. Wendt, F. Wienholtz, R. N. Wolf, A. Zadvornaya, and K. Zuber, “Shape staggering of mid-shell mercury isotopes from in-source laser spectroscopy compared with density functional theory and monte carlo shell model calculations,” *To be submitted to Physical Review C*, 2018.
- [3] L. P. Gaffney et al., “Studies of pear-shaped nuclei using accelerated radioactive beams,” *Nature*, vol. 497, pp. 199 EP –, May 2013. Article.
- [4] I. Tanihata, H. Hamagaki, O. Hashimoto, S. Nagamiya, Y. Shida, N. Yoshikawa, O. Yamakawa, K. Sugimoto, T. Kobayashi, D. Greiner, N. Takahashi, and Y. Nojiri, “Measurements of interaction cross sections

- and radii of he isotopes,” *Physics Letters B*, vol. 160, no. 6, pp. 380 – 384, 1985.
- [5] G. Ulm, S. K. Bhattacharjee, P. Dabkiewicz, G. Huber, H. J. Kluge, T. K hl, H. Lochmann, E. W. Otten, K. Wendt, S. A. Ahmad, W. Klempt, and R. Neugart, “Isotope shift of ^{182}Hg and an update of nuclear moments and charge radii in the isotope range ^{181}Hg – ^{206}Hg ,” *Zeitschrift f r Physik A Atomic Nuclei*, vol. 325, no. 3, pp. 247–259, 1986.
- [6] A. N. Andreyev, M. Huyse, P. Van Duppen, L. Weissman, D. Ackermann, J. Gerl, F. P. Hessberger, S. Hofmann, A. Kleinbohl, G. Munzenberg, S. Reshitko, C. Schlegel, H. Schaffner, P. Cagarda, M. Matos, S. Saro, A. Keenan, C. Moore, C. D. O’Leary, R. D. Page, M. Taylor, H. Kettunen, M. Leino, A. Lavrentiev, R. Wyss, and K. Heyde, “A triplet of differently shaped spin-zero states in the atomic nucleus ^{186}Pb ,” *Nature*, vol. 405, pp. 430–433, May 2000.
- [7] O. Sorlin and M.-G. Porquet, “Nuclear magic numbers: New features far from stability,” *Progress in Particle and Nuclear Physics*, vol. 61, no. 2, pp. 602 – 673, 2008.
- [8] K. Heyde and J. Wood, “Shape coexistence in atomic nuclei,” *Review of Modern Physics*, vol. 83, pp. 1467 – 1521, 2011.
- [9] S. R. Beane, E. Chang, S. Cohen, W. Detmold, H. W. Lin, K. Orginos, A. Parre o, M. J. Savage, and B. C. Tiburzi, “Magnetic moments of light nuclei from lattice quantum chromodynamics,” *Phys. Rev. Lett.*, vol. 113, p. 252001, Dec 2014.
- [10] E. Epelbaum, “Few-nucleon forces and systems in chiral effective field theory,” *Progress in Particle and Nuclear Physics*, vol. 57, no. 2, pp. 654 – 741, 2006.
- [11] A. Signoracci, T. Duguet, G. Hagen, and G. R. Jansen, “Ab initio bogoliubov coupled cluster theory for open-shell nuclei,” *Phys. Rev. C*, vol. 91, p. 064320, Jun 2015.
- [12] M. Kortelainen, J. McDonnell, W. Nazarewicz, P.-G. Reinhard, J. Sarich, N. Schunck, M. V. Stoitsov, and S. M. Wild, “Nuclear energy density optimization: Large deformations,” *Phys. Rev. C*, vol. 85, p. 024304, Feb 2012.
- [13] R. Ferrer, A. Barzakh, B. Bastin, R. Beerwerth, M. Block, P. Creemers, H. Grawe, R. de Groote, P. Delahaye, X. Fl chard, S. Franchoo, S. Fritzsche, L. P. Gaffney, L. Ghys, W. Gins, C. Granados, R. Heinke, L. Hijazi, M. Huyse, T. Kron, M. Kudryavtsev, Yuand Laatiaoui,

- N. Lecesne, M. Loiselet, F. Lutton, I. D. Moore, Y. Martínez, E. Mogilevskiy, P. Naubereit, J. Piot, S. Raeder, S. Rothe, H. Savajols, S. Sels, V. Sonnenschein, J.-C. Thomas, E. Traykov, C. Van Beveren, P. Van den Bergh, P. Van Duppen, K. Wendt, and A. Zadvornaya, “Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion,” *Nature Communications*, vol. 8, pp. 14520 EP –, Feb 2017. Article.
- [14] R. Heinke, T. Kron, S. Raeder, T. Reich, P. Schönberg, M. Trümper, C. Weichhold, and K. Wendt, “High-resolution in-source laser spectroscopy in perpendicular geometry,” *Hyperfine Interactions*, vol. 238, p. 6, Dec 2016.
- [15] S. Ettenauer et al., “Benchmarking of a multi ion reflection apparatus for collinear laser spectroscopy of radionuclides,” 2017.
- [16] T. Cocolios, H. A. Suradi, J. Billowes, I. Budinčević, R. de Groote, S. D. Schepper, V. Fedosseev, K. Flanagan, S. Franchoo, R. G. Ruiz, H. Heylen, F. L. Blanc, K. Lynch, B. Marsh, P. Mason, G. Neyens, J. Papuga, T. Procter, M. Rajabali, R. Rossel, S. Rothe, G. Simpson, A. Smith, I. Strashnov, H. Stroke, D. Verney, P. Walker, K. Wendt, and R. Wood, “The collinear resonance ionization spectroscopy (cris) experimental setup at cern-isolde,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 317, pp. 565 – 569, 2013. XVIth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 2–7, 2012 at Matsue, Japan.
- [17] P. Campbell, I. Moore, and M. Pearson, “Laser spectroscopy for nuclear structure physics,” *Progress in Particle and Nuclear Physics*, vol. 86, pp. 127 – 180, 2016.
- [18] Y. Blumenfeld, T. Nilsson, and P. V. Duppen, “Facilities and methods for radioactive ion beam production,” *Physica Scripta*, vol. 2013, no. T152, p. 014023, 2013.
- [19] “Atom-at-a-time laser resonance ionization spectroscopy of nobelium,” *Nature*, vol. 538, no. 7626, pp. 495–498, 2016.
- [20] S. Hofmann and G. Münzenberg, “The discovery of the heaviest elements,” *Rev. Mod. Phys.*, vol. 72, pp. 733–767, Jul 2000.
- [21] B. Fricke and K. Heilig, “Nuclear charge radii,” *Landolt-Bornstein*, vol. 20, 2004.

- [22] J. Bonn, J. Huber, H.-J. Kluge, and W. Otten, E., “Sudden change in the nuclear charge distribution of very light mercury isotopes,” *Physics Letters B*, vol. 38, pp. 308 – 311, 1972.
- [23] T. E. Cocolios, W. Dexters, M. D. Seliverstov, A. N. Andreyev, S. Antalic, A. E. Barzakh, B. Bastin, J. Büscher, I. G. Darby, D. V. Fedorov, V. N. Fedosseyev, K. T. Flanagan, S. Franchoo, S. Fritzsche, G. Huber, M. Huyse, M. Keupers, U. Köster, Y. Kudryavtsev, E. Mané, B. A. Marsh, P. L. Molkanov, R. D. Page, A. M. Sjoedin, I. Stefan, J. Van de Walle, P. Van Duppen, M. Venhart, S. G. Zemlyanoy, M. Bender, and P.-H. Heenen, “Early onset of ground state deformation in neutron deficient polonium isotopes,” *Phys. Rev. Lett.*, vol. 106, p. 052503, Feb 2011.
- [24] L. W. Wansbeek, S. Schlessers, B. K. Sahoo, A. E. L. Dieperink, C. J. G. Onderwater, and R. G. E. Timmermans, “Charge radii of radium isotopes,” *Phys. Rev. C*, vol. 86, p. 015503, Jul 2012.
- [25] J. Cubiss et al., “Charge radii and electromagnetic moments of 19x-211 at isotopes,” *In preparation*, 2017.
- [26] G. J. Farooq-Smith, T. E. Cocolios, J. Billowes, M. L. Bissell, I. Budinčević, T. Day Goodacre, R. P. de Groote, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, R. Li, K. M. Lynch, B. A. Marsh, G. Neyens, R. E. Rossel, S. Rothe, H. H. Stroke, K. D. A. Wendt, S. G. Wilkins, and X. F. Yang, “Laser and decay spectroscopy of the short-lived isotope ^{214}Fr in the vicinity of the $n = 126$ shell closure,” *Phys. Rev. C*, vol. 94, p. 054305, Nov 2016.
- [27] K. M. Lynch, S. G. Wilkins, J. Billowes, C. L. Binnersley, M. L. Bissell, K. Chrysalidis, T. E. Cocolios, T. D. Goodacre, R. P. de Groote, G. J. Farooq-Smith, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, W. Gins, R. Heinke, A. Koszorús, B. A. Marsh, P. L. Molkanov, P. Naubereit, G. Neyens, C. M. Ricketts, S. Rothe, C. Seiffert, M. D. Seliverstov, H. H. Stroke, D. Studer, A. R. Vernon, K. D. A. Wendt, and X. F. Yang, “Laser-spectroscopy studies of the nuclear structure of neutron-rich radium,” *Phys. Rev. C*, vol. 97, p. 024309, Feb 2018.
- [28] I. Angeli and K. Marinova, “Table of experimental nuclear ground state charge radii: An update,” *Atomic Data and Nuclear Data Tables*, vol. 99, no. 1, pp. 69 – 95, 2013.
- [29] M. D. Seliverstov, T. E. Cocolios, and W. Dexters et al., “Electromagnetic moments of odd- a $^{193}\text{~}^{203,211}\text{Po}$ isotopes,” *Phys. Rev. C*, vol. 89, p. 034323, Mar 2014.

- [30] A. E. Barzakh, D. V. Fedorov, V. S. Ivanov, P. L. Molkanov, F. V. Moroz, S. Y. Orlov, V. N. Panteleev, M. D. Seliverstov, and Y. M. Volkov, "Laser spectroscopy studies of intruder states in $^{193,195,197}\text{Bi}$," *Phys. Rev. C*, vol. 94, p. 024334, Aug 2016.
- [31] A. E. Barzakh, D. V. Fedorov, V. S. Ivanov, P. L. Molkanov, F. V. Moroz, S. Y. Orlov, V. N. Panteleev, M. D. Seliverstov, and Y. M. Volkov, "Onset of deformation in neutron-deficient bi isotopes studied by laser spectroscopy," *Phys. Rev. C*, vol. 95, p. 044324, Apr 2017.
- [32] A. E. Barzakh, L. K. Batist, D. V. Fedorov, V. S. Ivanov, K. A. Mezilev, P. L. Molkanov, F. V. Moroz, S. Y. Orlov, V. N. Panteleev, and Y. M. Volkov, "Changes in the mean-square charge radii and magnetic moments of neutron-deficient tl isotopes," *Phys. Rev. C*, vol. 88, p. 024315, Aug 2013.
- [33] A. E. Barzakh, A. N. Andreyev, T. E. Cocolios, R. P. de Groote, D. V. Fedorov, V. N. Fedosseev, R. Ferrer, D. A. Fink, L. Ghys, M. Huyse, U. Köster, J. Lane, V. Liberati, K. M. Lynch, B. A. Marsh, P. L. Molkanov, T. J. Procter, E. Rapisarda, S. Rothe, K. Sandhu, M. D. Seliverstov, A. M. Sjödin, C. Van Beveren, P. Van Duppen, M. Venhart, and M. Veselský, "Changes in mean-squared charge radii and magnetic moments of $^{179-184}\text{Tl}$ measured by in-source laser spectroscopy," *Phys. Rev. C*, vol. 95, p. 014324, Jan 2017.
- [34] K. Heyde and J. L. Wood, "Nuclear shapes: from earliest ideas to multiple shape coexisting structures," *Physica Scripta*, vol. 91, no. 8, p. 083008, 2016.
- [35] N. Rud, D. Ward, H. R. Andrews, R. L. Graham, and J. S. Geiger, "Lifetimes in the ground-state band of ^{184}Hg ," *Phys. Rev. Lett.*, vol. 31, pp. 1421–1423, Dec 1973.
- [36] D. Proetel, R. M. Diamond, P. Kienle, J. R. Leigh, K. H. Maier, and F. S. Stephens, "Evidence for strongly deformed shapes in ^{186}Hg ," *Phys. Rev. Lett.*, vol. 31, pp. 896–898, Oct 1973.
- [37] J. Bonn, G. Huber, H. J. Kluge, and E. W. Otten, "Spins, moments and charge radii in the isotopic series ^{181}hg - ^{191}hg ," *Zeitschrift für Physik A Atoms and Nuclei*, vol. 276, no. 3, pp. 203–217, 1976.
- [38] T. Kühn, P. Dabkiewicz, C. Duke, H. Fischer, H. J. Kluge, H. Kremmling, and E. W. Otten, "Nuclear shape staggering in very neutron-deficient hg isotopes detected by laser spectroscopy," *Phys. Rev. Lett.*, vol. 39, pp. 180–183, Jul 1977.

- [39] P. Dabkiewicz, F. Buchinger, H. Fischer, H.-J. Kluge, H. Kremmling, T. Kühl, A. Müller, and H. Schuessler, “Nuclear shape isomerism in ^{185}Hg detected by laser spectroscopy,” *Physics Letters B*, vol. 82, no. 2, pp. 199 – 203, 1979.
- [40] P. M. Goddard, P. D. Stevenson, and A. Rios, “Charge radius isotope shift across the $n=126$ shell gap,” *Phys. Rev. Lett.*, vol. 110, p. 032503, Jan 2013.
- [41] Goddard, P.M., Stevenson, P.D., and Rios, A., “Cause of the charge radius isotope shift at the $n=126$ shell gap,” *EPJ Web of Conferences*, vol. 66, p. 02042, 2014.
- [42] J.-P. Ebran, A. Mutschler, E. Khan, and D. Vretenar, “Spin-orbit interaction in relativistic nuclear structure models,” *Phys. Rev. C*, vol. 94, p. 024304, Aug 2016.
- [43] T. E. Cocolios, “A new perspective on charge radii around $z = 82$,” *Hyperfine Interactions*, vol. 238, p. 16, Jan 2017.
- [44] V. Somà, A. Cipollone, C. Barbieri, P. Navrátil, and T. Duguet, “Chiral two- and three-nucleon forces along medium-mass isotope chains,” *Phys. Rev. C*, vol. 89, p. 061301, Jun 2014.
- [45] M. Bender, P.-H. Heenen, and P.-G. Reinhard, “Self-consistent mean-field models for nuclear structure,” *Rev. Mod. Phys.*, vol. 75, pp. 121–180, Jan 2003.
- [46] P. Ring and P. Schuck, *The nuclear many-body problem*. Springer Science & Business Media, 2004.
- [47] W. D. Myers and W. Swiatecki, “Average nuclear properties,” *Annals of Physics*, vol. 55, no. 3, pp. 395 – 505, 1969.
- [48] W. Myers and W. Swiatecki, “The nuclear droplet model for arbitrary shapes,” *Annals of Physics*, vol. 84, no. 1, pp. 186 – 210, 1974.
- [49] D. A. Fink, T. E. Cocolios, A. N. Andreyev, S. Antalic, A. E. Barzakh, B. Bastin, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, L. Ghys, A. Gottberg, M. Huyse, N. Imai, T. Kron, N. Lemesne, K. M. Lynch, B. A. Marsh, D. Pauwels, E. Rapisarda, S. D. Richter, R. E. Rossel, S. Rothe, M. D. Seliverstov, A. M. Sjödin, C. Van Beveren, P. Van Duppen, and K. D. A. Wendt, “In-source laser spectroscopy with the laser ion source and trap: First direct study of the ground-state properties of $^{217,219}\text{Po}$,” *Phys. Rev. X*, vol. 5, p. 011018, Feb 2015.

- [50] B. et al., “Changes in mean-squared charge radii and magnetic moments of $^{179}\text{--}^{184}\text{Tl}$ measured by in-source laser spectroscopy,” *Phys. Rev. C*, vol. 95, p. 014324, Jan 2017.
- [51] W. D. Myers and K.-H. Schmidt, “An update on droplet-model charge distributions,” *Nuclear Physics A*, vol. 410, no. 1, pp. 61 – 73, 1983.
- [52] D. Berdichevsky and F. Tondeur, “Nuclear core densities, isotope shifts, and the parametrization of the droplet model,” *Zeitschrift für Physik A Atoms and Nuclei*, vol. 322, no. 1, pp. 141–147, 1985.
- [53] E. W. Otten, *Nuclear Radii and Moments of Unstable Isotopes*, pp. 517–638. Boston, MA: Springer US, 1989.
- [54] P. Möller, W. Myers, W. Swiatecki, and J. Treiner, “Nuclear mass formula with a finite-range droplet model and a folded-yukawa single-particle potential,” *Atomic Data and Nuclear Data Tables*, vol. 39, no. 2, pp. 225 – 233, 1988.
- [55] M. Goeppert-Mayer, *The Shell Model, Nobel lecture in physics 1963-1970*. Elsevier Publishing Company, Amsterdam, 1972.
- [56] K. Krane, *Introductory Nuclear Physics*. Wiley, 1987.
- [57] K. Heyde, *The Nuclear Shell Model*. Springer-Verlag, Berlin, 1994.
- [58] V. Strutinsky, “Shell effects in nuclear masses and deformation energies,” *Nuclear Physics A*, vol. 95, no. 2, pp. 420 – 442, 1967.
- [59] R. Casten and R. Casten, *Nuclear Structure from a Simple Perspective*. Oxford science publications, Oxford University Press, 2000.
- [60] H. Grawe, *Shell Model from a Practitioner’s Point of View*, pp. 33–75. Berlin, Heidelberg: Springer Berlin Heidelberg, 2004.
- [61] P. Van Isacker and S. Pittel, “Symmetries and deformations in the spherical shell model,” *Physica Scripta*, vol. 91, no. 2, p. 023009, 2016.
- [62] T. Otsuka and Y. Tsunoda, “The role of shell evolution in shape coexistence,” *Journal of Physics G: Nuclear and Particle Physics*, vol. 43, no. 2, p. 024009, 2016.
- [63] J. E. García-Ramos and K. Heyde, “Nuclear shape coexistence: A study of the even-even hg isotopes using the interacting boson model with configuration mixing,” *Phys. Rev. C*, vol. 89, p. 014306, Jan 2014.

- [64] K. Nomura, R. Rodríguez-Guzmán, and L. M. Robledo, “Shape evolution and the role of intruder configurations in hg isotopes within the interacting boson model based on a gogny energy density functional,” *Phys. Rev. C*, vol. 87, p. 064313, Jun 2013.
- [65] J. M. Yao, M. Bender, and P.-H. Heenen, “Systematics of low-lying states of even-even nuclei in the neutron-deficient lead region from a beyond-mean-field calculation,” *Phys. Rev. C*, vol. 87, p. 034322, Mar 2013.
- [66] N. Shimizu, T. Abe, M. Honma, T. Otsuka, T. Togashi, Y. Tsunoda, Y. Utsuno, and T. Yoshida, “Monte carlo shell model studies with massively parallel supercomputers,” *Physica Scripta*, vol. 92, no. 6, p. 063001, 2017.
- [67] T. Otsuka, M. Honma, T. Mizusaki, N. Shimizu, and Y. Utsuno, “Monte carlo shell model for atomic nuclei,” *Progress in Particle and Nuclear Physics*, vol. 47, no. 1, pp. 319 – 400, 2001.
- [68] S. Nilsson, “Deformed potential shell model,” *Mat. Fys. Medd. Dan. Vid/Selsk.*, vol. 29, no. 16, 1955.
- [69] R. B. Firestone and V. S. Shirley, “Table of isotopes, 2 volume set,” *Table of Isotopes, 2 Volume Set, by Richard B. Firestone, Virginia S. Shirley (Editor), pp. 3168. ISBN 0-471-33056-6. Wiley-VCH, December 1998.*, p. 3168, 1998.
- [70] T. Nikšić, D. Vretenar, and P. Ring, “Relativistic nuclear energy density functionals: Mean-field and beyond,” *Progress in Particle and Nuclear Physics*, vol. 66, no. 3, pp. 519 – 548, 2011.
- [71] D. L. Hill and J. A. Wheeler, “Nuclear constitution and the interpretation of fission phenomena,” *Phys. Rev.*, vol. 89, pp. 1102–1145, Mar 1953.
- [72] H.-J. Kluge and W. Nörtershäuser, “Lasers for nuclear physics,” *Spectrochimica Acta Part B: Atomic Spectroscopy*, vol. 58, no. 6, pp. 1031 – 1045, 2003.
- [73] J. Billowes and P. Campbell, “High-resolution laser spectroscopy for the study of nuclear sizes and shapes,” *Journal of Physics G: Nuclear and Particle Physics*, vol. 21, no. 6, p. 707, 1995.
- [74] G. Neyens, “Nuclear magnetic and quadrupole moments for nuclear structure research on exotic nuclei,” *Reports on Progress in Physics*, vol. 66, no. 4, p. 633, 2003.

- [75] K. Blaum, J. Dilling, and W. Nörtershäuser, “Precision atomic physics techniques for nuclear physics with radioactive beams,” *Phys. Scr.*, vol. T152, p. 014017, Jan 2013.
- [76] N. J. Stone, “New table of recommended nuclear electric quadrupole moments,” *Hyperfine Interactions*, vol. 230, no. 1, pp. 7–16, 2015.
- [77] J. E. Rosenthal and G. Breit, “The isotope shift in hyperfine structure,” *Phys. Rev.*, vol. 41, pp. 459–470, Aug 1932.
- [78] A. Bohr and V. F. Weisskopf, “The influence of nuclear structure on the hyperfine structure of heavy elements,” *Phys. Rev.*, vol. 77, pp. 94–98, Jan 1950.
- [79] J. Persson, “Table of hyperfine anomaly in atomic systems,” *Atomic Data and Nuclear Data Tables*, vol. 99, no. 1, pp. 62 – 68, 2013.
- [80] E. C. Seltzer, “ k x-ray isotope shifts,” *Phys. Rev.*, vol. 188, pp. 1916–1919, Dec 1969.
- [81] W. Nörtershäuser, G. C., C. Scheidenberger, and M. Pfützner, *The Euroscool on Exotic Beams, Ch.6: nuclear charge radii of light elements and recent developments*, vol. 4. Springer, 2014.
- [82] W. Demtröder, *Laser spectroscopy: basic concepts and instrumentation*. Springer Science & Business Media, 2013.
- [83] P. R. Berman and V. S. Malinovsky, *Principles of laser spectroscopy and quantum optics*. Princeton University Press, 2010.
- [84] “Nist atomic level database,” 2018.
- [85] Y. Kudryavtsev, R. Ferrer, M. Huyse, P. V. den Bergh, and P. V. Duppen, “The in-gas-jet laser ion source: Resonance ionization spectroscopy of radioactive atoms in supersonic gas jets,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 297, pp. 7 – 22, 2013.
- [86] R. P. de Groote, M. Verlinde, V. Sonnenschein, K. T. Flanagan, I. Moore, and G. Neyens, “Efficient, high-resolution resonance laser ionization spectroscopy using weak transitions to long-lived excited states,” *Phys. Rev. A*, vol. 95, p. 032502, Mar 2017.
- [87] M. J. G. Borge and K. Blaum, “Focus on exotic beams at isolde: A laboratory portrait,” *Journal of Physics G: Nuclear and Particle Physics*, vol. 45, no. 1, p. 010301, 2018.

- [88] T. Day Goodacre et al., “Blurring the boundaries between ion sources: The application of the rilis inside a febiad type ion source at isolde,” *Nuclear Instrumentation and Methods in Physcs Research B*, vol. 376, pp. 39 – 45, 2016.
- [89] “Isolde website, <http://isolde.web.cern.ch/>,” 2017.
- [90] T. Day Goodacre, J. Billowes, K. Chrysalidis, D. V. Fedorov, V. N. Fedosseev, B. A. Marsh, P. L. Molkanov, R. E. Rossel, S. Rothe, C. Seiffert, and K. D. A. Wendt, “Rilis-ionized mercury and tellurium beams at isolde cern,” *Hyperfine Interactions*, vol. 238, p. 41, Feb 2017.
- [91] J. Elseviers, A. N. Andreyev, M. Huyse, P. Van Duppen, S. Antalic, A. Barzakh, N. Bree, T. E. Cocolios, V. F. Comas, J. Diriken, D. Fedorov, V. N. Fedosseev, S. Franchoo, L. Ghys, J. A. Heredia, O. Ivanov, U. Köster, B. A. Marsh, K. Nishio, R. D. Page, N. Patronis, M. D. Seliverstov, I. Tsekhanovich, P. Van den Bergh, J. Van De Walle, M. Venhart, S. Vermote, M. Veselský, and C. Wagemans, “ β -delayed fission of ^{180}tl ,” *Phys. Rev. C*, vol. 88, p. 044321, Oct 2013.
- [92] B. Lommel, H. W., K. B., and S. J., “Preparation of self-supporting carbon thin films,” *Nuclear Instrumentation and Methods in Physcs Research A*, vol. 480, pp. 199 – 203, 2002.
- [93] J. Cubiss, “Phd thesis university of york: In-source laser spectroscopy of at isotopes and decay studies of ^{178}au ,” 2017.
- [94] R. N. Wolf et al., “Isoltrap’s multi-reflection time-of-flight mass separator/spectrometer,” *International Journal of Mass Spectrometry*, vol. 349-350, pp. 123–133, 2013.
- [95] “Brookhaven national nuclear data center (nnrc) - evaluated nuclear structure data file (ensdf).”
- [96] W. Gins, R. de Groote, M. Bissell, C. G. Buitrago, R. Ferrer, K. Lynch, G. Neyens, and S. Sels, “Analysis of counting data: Development of the satlas python package,” *Computer Physics Communications*, vol. 222, no. Supplement C, pp. 286 – 294, 2018.
- [97] M. Newville, T. Stensitzki, D. B. Allen, and A. Ingargiola, “LMFIT: Non-Linear Least-Square Minimization and Curve-Fitting for Python,” sep 2014.
- [98] R. Brun and F. Rademakers, “Root - an object oriented data analysis framework,” *Proceedings AINHENP’96 Workshop, Lausanne, Nucl. Inst. and Meth. in Phys. Res. A*, vol. 389, pp. 81 – 86, 1997.

- [99] S. Raman, C. Nestor, and P. Tikkanen, "Transition probability from the ground to the first-excited $2+$ state of even-even nuclides," *Atomic Data and Nuclear Data Tables*, vol. 78, no. 1, pp. 1 – 128, 2001.
- [100] R. J. Reimann and M. N. McDermott, "Precision magnetic moment determinations for 43-min ^{199m}Hg and other isomers of mercury," *Phys. Rev. C*, vol. 7, pp. 2065–2079, May 1973.
- [101] P. Moskowitz and M. Lombardi, "Distribution of nuclear magnetization in mercury isotopes," *Physics Letters B*, vol. 46, no. 3, pp. 334 – 336, 1973.
- [102] A.-M. Mårtensson-Pendrill, "Magnetic moment distributions in tl nuclei," *Phys. Rev. Lett.*, vol. 74, pp. 2184–2187, Mar 1995.
- [103] H. J. Rosenberg and H. H. Stroke, "Effect of a diffuse nuclear charge distribution on the hyperfine-structure interaction," *Phys. Rev. A*, vol. 5, pp. 1992–2000, May 1972.
- [104] T. Fujita and A. Arima, "Magnetic hyperfine structure of muonic and electronic atoms," *Nuclear Physics A*, vol. 254, no. 2, pp. 513 – 541, 1975.
- [105] F. Kondev, M. Carpenter, R. Janssens, C. Lister, and K. A. S. et al., "First observation of excited structures in neutron-deficient ^{179}Hg : evidence for multiple shape coexistence," *Physics Letters B*, vol. 528, no. 3–4, pp. 221 – 227, 2002.
- [106] D. G. Jenkins, A. N. Andreyev, R. D. Page, M. P. Carpenter, R. V. F. Janssens, C. J. Lister, and F. G. Kondev et al., "Confirmation of triple shape coexistence in ^{179}Hg : focal plane spectroscopy of the α decay of ^{183}Pb ," *Phys. Rev. C*, vol. 66, p. 011301, Jul 2002.
- [107] M. Anselment, W. Faubel, S. Göring, A. Hanser, G. Meisel, H. Rebel, and G. Schatz, "The odd-even staggering of the nuclear charge radii of pb isotopes," *Nuclear Physics A*, vol. 451, no. 3, pp. 471 – 480, 1986.
- [108] R. H. Kohler, "Detection of double resonance by frequency change: Application to hg^{201} ," *Phys. Rev.*, vol. 121, pp. 1104–1111, Feb 1961.
- [109] J. Bieroń, P. Pyykkö, and P. Jönsson, "Nuclear quadrupole moment of ^{201}Hg ," *Phys. Rev. A*, vol. 71, p. 012502, Jan 2005.
- [110] T. Togashi et al., "Quantum phase transition in the shape of zr isotopes," *Phys. Rev. Lett.*, vol. 117, p. 172502, Oct 2016.
- [111] S. Suchyta et al., "Shape coexistence in ^{68}Ni ," *Phys. Rev. C*, vol. 89, p. 021301, Feb 2014.

- [112] S. Hofmann and G. Münzenberg, "The discovery of the heaviest elements," *Rev. Mod. Phys.*, vol. 72, pp. 733–767, Jul 2000.
- [113] A. Sobiczewski and K. Pomorski, "Description of structure and properties of superheavy nuclei," *Progress in Particle and Nuclear Physics*, vol. 58, no. 1, pp. 292 – 349, 2007.
- [114] Y. T. Oganessian and V. K. Utyonkov, "Super-heavy element research," *Reports on Progress in Physics*, vol. 78, no. 3, p. 036301, 2015.
- [115] Y. T. Oganessian, V. K. Utyonkov, Y. V. Lobanov, F. S. Abdullin, A. N. Polyakov, R. N. Sagaidak, I. V. Shirokovsky, Y. S. Tsyganov, A. A. Voinov, G. G. Gulbekian, S. L. Bogomolov, B. N. Gikal, A. N. Mezentsev, S. Iliev, V. G. Subbotin, A. M. Sukhov, K. Subotic, V. I. Zagrebaev, G. K. Vostokin, M. G. Itkis, K. J. Moody, J. B. Patin, D. A. Shaughnessy, M. A. Stoyer, N. J. Stoyer, P. A. Wilk, J. M. Keneally, J. H. Landrum, J. F. Wild, and R. W. Loughheed, "Synthesis of the isotopes of elements 118 and 116 in the ^{249}Cf and $^{245}\text{Cm} + ^{48}\text{Ca}$ fusion reactions," *Phys. Rev. C*, vol. 74, p. 044602, Oct 2006.
- [116] L. Öhrström and J. Reedijk, *pac*, vol. 88, ch. Names and symbols of the elements with atomic numbers 113, 115, 117 and 118 (IUPAC Recommendations 2016), p. 1225. 2018 2016. 12.
- [117] L. Vermeeren, N. Bijmens, M. Huyse, Y. A. Kudryavtsev, P. Van Duppen, J. Wauters, Z. N. Qamhieh, P. Thoen, E. Vandeweert, and R. E. Silverans, "An on-line laser ion source based on resonance photoionization in a gas cell," *Phys. Rev. Lett.*, vol. 73, pp. 1935–1938, Oct 1994.
- [118] Y. Kudryavtsev, J. Andrzejewski, N. Bijmens, S. Franchoo, J. Gentens, M. Huyse, A. Piechaczek, J. Szerypo, I. Reusen, P. V. Duppen, P. V. D. Bergh, L. Vermeeren, J. Wauters, and A. Wöhr, "Beams of short lived nuclei produced by selective laser ionization in a gas cell," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 114, no. 3, pp. 350 – 365, 1996.
- [119] T. E. Cocolios, A. N. Andreyev, B. Bastin, N. Bree, J. Büscher, J. Elseviers, J. Gentens, M. Huyse, Y. Kudryavtsev, D. Pauwels, T. Sonoda, P. Van den Bergh, and P. Van Duppen, "Magnetic dipole moment of $^{57,59}\text{Cu}$ measured by in-gas-cell laser spectroscopy," *Phys. Rev. Lett.*, vol. 103, p. 102501, Aug 2009.
- [120] Y. Kudryavtsev, P. Creemers, R. Ferrer, C. Granados, L. Gaffney, M. Huyse, E. Mogilevskiy, S. Raeder, S. Sels, P. V. den Bergh,

- P. V. Duppen, and A. Zadornaya, “A new in-gas-laser ionization and spectroscopy laboratory for off-line studies at ku leuven,” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 376, no. Supplement C, pp. 345 – 352, 2016. Proceedings of the XVIIth International Conference on Electromagnetic Isotope Separators and Related Topics (EMIS2015), Grand Rapids, MI, U.S.A., 11-15 May 2015.
- [121] R. P. de Groote, I. Budinčević, J. Billowes, M. L. Bissell, T. E. Cocolios, G. J. Farooq-Smith, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, R. Li, K. M. Lynch, B. A. Marsh, G. Neyens, R. E. Rossel, S. Rothe, H. H. Stroke, K. D. A. Wendt, S. G. Wilkins, and X. Yang, “Use of a continuous wave laser and pockels cell for sensitive high-resolution collinear resonance ionization spectroscopy,” *Phys. Rev. Lett.*, vol. 115, p. 132501, Sep 2015.
- [122] H. Dehmelt, “Radiofrequency spectroscopy of stored ions,” vol. 3 of *Advances in Atomic and Molecular Physics*, pp. 53 – 72, Academic Press, 1968.
- [123] G. Bollen, “Traps for rare isotopes,” in *The Euroschool Lectures on Physics with Exotic Beams, Vol. I* (J. Al-Khalili and E. Roeckl, eds.), vol. 651 of *Lecture Notes in Physics*, pp. 169 –210, Springer Berlin Heidelberg, 2004.
- [124] P. Miller and M. Bonner Denton, “The transmission properties of an rf-only quadrupole mass filter,” vol. 72, p. 223, 1986.
- [125] A. Kellerbauer, T. Kim, R. Moore, and P. Varfalvy, “Buffer gas cooling of ion beams,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 469, no. 2, pp. 276 – 285, 2001.
- [126] S. Schwarz, “Ioncool—a versatile code to characterize gas-filled ion bunchers and coolers (not only) for nuclear physics applications,” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 566, no. 2, pp. 233 – 243, 2006.
- [127] S. Schwarz, “Simulations for ion traps – buffer gas cooling,” in *The Euroschool Lectures on Physics with Exotic Beams, Vol. I* (K. Blaum and F. Herfurt, eds.), *Lecture Notes in Physics*, pp. 97 –116, Springer Berlin Heidelberg, 2008.
- [128] J. Moore, C. Davis, and M. E. Coplan, *Building scientific apparatus*. Cambridge University Press, 2009.

- [129] S. Schwarz, G. Bollen, D. Lawton, A. Neudert, R. Ringle, P. Schury, and T. Sun, "A second-generation ion beam buncher and cooler," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 204, pp. 474 – 477, 2003. 14th International Conference on Electromagnetic Isotope Separators and Techniques Related to their Applications.
- [130] G. Bollen et al, "Beam cooling at the low-energy-beam and ion-trap facility at nscl/msu," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 532, no. 1–2, pp. 203 – 209, 2004. International Workshop on Beam Cooling and Related Topics.
- [131] D. Manura and D. Dahl, "Simion (r) 8.0 user manual," January 2008.
- [132] D. Manura, "Additional notes on the simion hs1 collision model," *Included in SIMION software examples*, 2007.
- [133] "Comsol multiphysics, <https://www.comsol.com/>."
- [134] A. Zadvornaya, "Phd thesis ku leuven: Characterization of the spectral resolution of the in-gas-jet laser ionization spectroscopy method."
- [135] "Webelements database, <https://www.webelements.com/>."
- [136] X.-F. Tong, C.-L. Yang, M.-S. Wang, X.-G. Ma, and D.-H. Wang, "Interactions of $mz-x$ complexes ($m = cu, ag, and au; x = he, ne, and ar; and z = \pm 1$)," *The Journal of Chemical Physics*, vol. 134, no. 2, p. 024306, 2011.
- [137] F. Paschen, "Ueber die zum funkenübergang in luft, wasserstoff und kohlendäure bei verschiedenen drucken erforderliche potentialdifferenz," *Annalen der Physik*, vol. 273, no. 5, pp. 69–96, 1889.
- [138] M. A. Liebermann and A. J. Lichtenberg, *Principles of plasma discharges and materials processing*. Wiley, 2 ed., April 2005.
- [139] H. B. Smith, C. Charles, and R. W. Boswell, "Breakdown behavior in radio-frequency argon discharges," *Physics of Plasmas*, vol. 10, no. 3, pp. 875–881, 2003.
- [140] K. T. McDonald and D. P. Russell, "Methods of emittance measurement," *Frontiers of Particle Beams; Observation, Diagnosis and Correction*, pp. 122–132, 1989.
- [141] B. Barquest, "Phd thesis, michigan state university: An advanced ion guide for beam cooling and bunching for collinear laser spectroscopy of rare isotopes," 2014.

- [142] R. Ferrer, A. Kwiatkowski, G. Bollen, D. Lincoln, D. Morrissey, G. Pang, R. Ringle, J. Savory, and S. Schwarz, "Ion beam properties after mass filtering with a linear radiofrequency quadrupole," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 735, pp. 382 – 389, 2014.
- [143] Y. Kudryavtsev, B. Bruyneel, M. Huyse, J. Gentens, P. V. den Bergh, P. V. Duppen, and L. Vermeeren, "A gas cell for thermalizing, storing and transporting radioactive ions and atoms. part i: Off-line studies with a laser ion source," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 179, no. 3, pp. 412 – 435, 2001.
- [144] Y. Luo, *Comprehensive Handbook of Chemical Bond Energies*. CRC Press, Boca Raton, FL, 2007.
- [145] K. Wallmeroth, G. Bollen, A. Dohn, P. Egelhof, J. Grüner, F. Lindenlauf, U. Krönert, J. Campos, A. Rodriguez Yunta, M. J. G. Borge, A. Venugopalan, J. L. Wood, R. B. Moore, and H. J. Kluge, "Sudden change in the nuclear charge distribution of very light gold isotopes," *Phys. Rev. Lett.*, vol. 58, pp. 1516–1519, Apr 1987.
- [146] D. Verney, "Deformation change in light iridium nuclei from laser spectroscopy," *The European Physical Journal A - Hadrons and Nuclei*, vol. 30, pp. 489–518, Dec 2006.
- [147] R. Catherall, W. Andreazza, M. Breitenfeldt, A. Dorsival, G. J. Focker, T. P. Gharsa, G. T. J. J.-L. Grenard, F. Locci, P. Martins, S. Marzari, J. Schipper, A. Shornikov, and T. Stora, "The isolde facility," *Journal of Physics G: Nuclear and Particle Physics*, vol. 44, no. 9, p. 094002, 2017.
- [148] T. Stora and M. Kowalska, "Presentation: Proton energy increase to 2 gev: upgraded psb or rcs?," 2018.
- [149] N. Bree, K. Wrzosek-Lipska, and P. et al., "Shape coexistence in the neutron-deficient even-even $^{182-188}\text{Hg}$ isotopes studied via coulomb excitation," *Phys. Rev. Lett.*, vol. 112, p. 162701, Apr 2014.
- [150] S. J. Freeman *et al.*, "Letter of Intent to the ISOLDE and Neutron Time-of-Flight Experiments Committee for experiments with HIE-ISOLDE: A HELIcal Orbit Spectrometer (HELIOS) for HIE-ISOLDE," Tech. Rep. CERN-INTC-2010-031. INTC-I-099, CERN, Geneva, May 2010.
- [151] B. Fernandez-Dominguez *et al.*, "Single particle structure of exotic nuclei with transfer reactions," *Progress in Particle and Nuclear Physics*, vol. 59, no. 1, pp. 389 – 391, 2007. International Workshop on Nuclear Physics 28th Course.

- [152] K. Tsukada, A. Enokizono, T. Ohnishi, K. Adachi, T. Fujita, M. Hara, M. Hori, T. Hori, S. Ichikawa, K. Kurita, K. Matsuda, T. Suda, T. Tamae, M. Togasaki, M. Wakasugi, M. Watanabe, and K. Yamada, “First elastic electron scattering from ^{132}Xe at the scrit facility,” *Phys. Rev. Lett.*, vol. 118, p. 262501, Jun 2017.
- [153] B. Cagnac, “Doppler-free two-photon spectroscopy,” *Hyperfine Interactions*, vol. 24, pp. 19–41, Aug 1985.
- [154] R. Ruiz and K. Chrysalidis *et al*, “Exploring the feasibility of production and laser spectroscopy experiments of exotic silicon isotopes,” *CERN INTC I-176 LOI*, 2017.
- [155] A. Jokinen, “The jyvässkylä accelerator laboratory,” *Nuclear Physics News*, vol. 24, no. 4, pp. 4–10, 2014.
- [156] M. Lewitowicz, “The spiral 2 project,” *AIP Conference Proceedings*, vol. 891, no. 1, pp. 91–98, 2007.

FACULTY OF SCIENCE
DEPARTMENT OF PHYSICS AND ASTRONOMY
NUCLEAR SPECTROSCOPY GROUP

Celestijnenlaan 200D box 2418

B-3001 Leuven

simon.sels@kuleuven.be

www.fys.kuleuven.be/iks/ns/home

