

Exploring the magical tenacity of doubly closed-core nuclei with gallium and francium isotopes

CERN-THESIS-2019-188
11/10/2019



Gregory James Farooq-Smith

Supervisor:
Prof. dr. T. E. Cocolios

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

October 2019

Exploring the magical tenacity of doubly closed-core nuclei with gallium and francium isotopes

Gregory James Farooq-Smith

Examination committee:

Prof. dr. A. Vantomme, chair
Prof. dr. T. E. Cocolios, supervisor
Prof. dr. P. Van Duppen
Prof. dr. G. Neyens
Prof. dr. R. Raabe

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

Dr. B. Cheal
(University of Liverpool, UK)
Dr. D. Verney
(IPN Orsay, France)

October 2019

© 2019 KU Leuven – Faculty of Science

Uitgegeven in eigen beheer, Gregory James Farooq-Smith , 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.

Acknowledgements

On the day I found out I would soon be starting a Ph.D in nuclear physics, I was at the BBC R1 studio in Broadcasting House in London, expressing my sheer disappointment upon the recent discovery of a few grey hairs. Now almost five-and-a-half years later, and possessing a larger tranche of grey hair, the time has come to reap the benefits of this rewarding experience by extending my already lengthy name by two extra letters: ‘Dr’. While I will not lie by saying that I have seen my fair share of challenges, I consider myself fabulously lucky to know such a smörgåsbord of people who have all immeasurably enriched my life. Therefore, the first few written pages of my thesis will now be dedicated to you all, because not doing so would amount to a level of absurdity far greater than the current state of Brexit affairs. For my larger groups of friends, you are mentioned in Table 1 below; I apologise profusely if I have missed anybody out.

I would first like to thank my supervisor, and also good friend: Thomas. You have given me the research opportunity others would simply dream of. Both professionally and socially, I have had the best of both worlds. I have learnt a wealth of knowledge from you, from discovering the beautiful intricacies of nuclear structure, to expanding my hitherto almost non-existent knowledge of French politics. I am especially thankful for all the enthusiasm, patience, and faith you have invested in me, which I truly believe has helped to shape the way I am today. I very much hope we keep in contact; at the very least, I need to know when you finish making your bed spread! I would also like to thank the other members of my jury (in alphabetical order): André, Bradley, David, Gerda, Piet, and Riccardo. It has been a pleasure to interact with you all during our numerous nuclear-physics discussions. I also thank my jury committee in general for providing me with valuable thesis feedback and suggestions, helping me to develop the Ph.D story I am truly proud to be telling.

My next acknowledgement is dedicated to the people I met where it all originally began for me: in Manchester, my spiritual home. I give the warmest of thank yous to all my past colleagues at The University of Manchester that helped me

Table 1: A table containing people whom I wish to thank for making my Ph.D journey so memorable. Excluding those part of ‘The CRIS collaboration’, people are grouped according to the region in which I first interacted with them.

Association	People
The CRIS collaboration.	Adam V., Agi, Ben, Chris R., Cory, Fredrik, Gerda, Henry S., Ivan B., Jon Bi., Kara, Kieran, Klaus, Mark, Ronald, Ruben, Serge, Shane, Thomas, Tobias, Wouter G., Xiao Fei.
The University of Manchester, and the Manchester region.	Adelle, Alex D., Alex G., Chris O., David R., Duncan H., Emily H., Faye, Henry C., Jack, James R., James G., Johnny, Jon Bi., Kieran, Liam V., Matt C., Mike G., Mike Ma., Mike Mu., Neha, Nic, Philip, Rhys, Roddy, Ruchi, Sam, Sarah C., Sarah E., Sean, Shane, Sharpey, Stephen, Stu, Thomas, Vinnie.
CERN, ISOLDE, LTA, and the Saint-Genis-Pouilly and Geneva regions.	Adam B., Adam P., Adam V., Amal, Andrei, Anna, Bijan, Boris, Bruce, Calvin, Cameron, Chloe, Daniel C., David L., Dinko, Duncan L., Ellis, Emily G., Emma, Erwin, Fenton, Frank, Hanne, Harry, Iwan, James C., Jon Bu., Kara, Kārlis, Katerina, Laurie, Liam G., Liam M., Lina, Magda, Mark, Matt B., Matus B., Matus S., Maxime M., Murdo, Razvan, Rebecca, Rob, Rose, Sarina, Sebastian, Stoyan, Sun, Tim, Tom B., Tom D. G., Vladimir, Vladislav, Will, Xanthe.
KU Leuven, and the Leuven region.	Agi, Ahmed, Alice, Andrés, Asteri, Bertrand, Camilo, Carlos, Charlotte, Christine, Cobus, Daniel, Dragan, Eline, Elisabeth, Elise, Eva, Gavin, Gertjan, Hannelore, Harsh, Ivan M., Ivan P., James McN., Janni, Jekabs, Jeroen, Jiecheng, Johanna, Karl, Kateryna, Koen, Kseniia, Leendert, Lennert, Lino, Liz, Manisha, Marek, Mateus, Matthias, Maxime R., Maya, Melinda, Nuno, Oleksii, Pepijn, Pin-Cheng, Renan, Ronald, Ruben, Sandro, Sasha, Simon Se., Simon St., Simon Va., Stefano, Steven, Takaharu, Tassos, Tiago, Valérie, Vera, Victoria, Vincent, Wouter G., Wouter R., Xiao Fei, Xu, Yisel.

take my first foray into nuclear physics. In particular to Kieran, my previous co-supervisor, who provided me with countless pearls of wisdom. A special mention also goes to the Nuclear Physics Drongos group, for the countless: crossbow-dart games, Guardian-quiz competitions, and not-so-occasional after-work drinks at the Sandbar. To everybody I met at the STFC nuclear-physics summer school in Lancaster, it was fun experiencing the synergy between physics and gin, as well as visiting Hustle three nights in a row. I also enjoyed the company of my housemates for the year I spent at Manchester: Matt, Rhys, and Shane. Watching back-to-back Premier League matches at the weekend, including the (almost) weekly £5 risk-free online gambling, were all thoroughly entertaining. To all of you whom I partied with at The Warehouse Project, Albert Hall, Factory *et al.*, thank you for the sensational memories together within (indisputably) one of the best party cities in the world.

I have also been incredibly lucky to have been based at one of the biggest physics laboratories in the world for almost a year and a half: CERN. Working within the nuclear-physics division, at the ISOLDE facility, I want to thank everybody for not only letting me collaborate with some of the most cutting-edge nuclear-physics research to date, but also all the best banter during lunchtimes in building 508. Particular recognition goes to my colleagues and friends at the CRIS setup, who have showed me all the wonderfully-fun aspects of research. It has been truly marvellous to witness the continual development of the CRIS setup into the pioneering experiment that it is today. Getting to know all the other UK LTA people that came to CERN made living in Saint-Genis-Pouilly that extra bit enjoyable. A special mention goes to the ‘Department of Mysteries’ (and its sequel: Electric Boogaloo) group, thank you for all: the exquisite recreational activities, the kebabs at Meyrin, the nightclubbing in L’Usine, and our ‘work outing’ to Amsterdam. These are memories I will cherish forever. To my housemates at ‘The Party House’: Jon, Laurie, Shane, and Stoyan, I enjoyed coming back home for all the ‘How I Met Your Mother’ marathons. We also really did throw the most kick-ass of parties!

Beginning my KU Leuven affiliation in 2015, I remember feeling somewhat like a lost puppy. Collectively, people at the IKS quickly made me feel part of the group for which I could not more thankful, despite (still) puzzling some of you with my audacious fashion choices. You also all taught me my most valuable life skill: how to speak good and understandable English. I also want to thank the secretaries Danielle, Fabienne, and Isabelle, and the IT support guys Bert and Luc, for their valuable assistance and support. To the (past and present) members of the Interdisciplinary Research group: Charlotte, Elisso, Hannelore, João, Kristof, Numa, Simon, Thomas, and Yisel, it has been wonderful interacting and working with you all, especially under the monikers COSYlicious and CERNtastic. A very special mention goes to the ‘da Love

Boat' group: Ivan, Koen, Leendert, Simon, and Tiago. Thank you for the copious: drinks and Bickies at the Oude Markt, board-game nights with *shuffle s'il vous plaît*, and €2 gambles at Metafoor, whose vowels I always accentuate. I very much look forward to our post-Ph.D meetups and continuing our activities for years to come! Also to my Leuven housemates, Eline and Liam, it has been so much fun binge-watching Netflix, drinking and *goeie babbels* at Café Belge and De Libertad, and cooking the most extravagant meals which have vastly improved my cooking repertoire. And of course, it has been lovely living with two of the fluffiest cats I will probably ever meet: Motoko and Plato.

Finally, I am so lucky my family have provided such an abundance of support throughout my every single step, despite my jet-setting ways across Europe. Mum and Dad, your unconditional support and love have helped me to get through my not-so-cheery moments during my studies. I have enjoyed so much whenever you have both visited me wherever I am, and it makes me so content to know that what I do, makes you both so proud of me. To my Nan, thank you for all of our chats, discussions, and ramblings whenever I have come to visit you, as well as all your unwavering support and love. I will definitely make sure I give you a copy of my thesis! Benedict and Rebekah, my brother and sister, it is always such a delight seeing your banterous messages pop up in our group chats. Although we do not see each other as often as we would like to, I have felt your support throughout my journey and you are both undoubtedly the best siblings I could have wished for. To Laura and Matt, my old childhood friends from secondary school, thank you both for your continual support and chats throughout the years. To think almost a decade has passed by so quickly since school! And Alison, as a teacher I have no doubt you will exercise meticulous vigilance for spelling mistakes in my thesis. This, along with countless other reasons, silly ways, mannerisms, and unique senses of humour, is what I love you so much for. Your immense support, patience, and enthusiasm have all kept me going at my lowest moments, while additionally keeping me sane and insane in equivalent measures. I am also so glad I convinced you that Leuven was not just some pretend, fairytale town. Now, I cannot wait to come back to you so we can start our lives together, along with our golden friends Woofles and Juni.

Cheers guys!

Greg



Abstract

This thesis presents nuclear spins, electromagnetic moments, and changes in the mean-square charge radii for radioactive gallium isotopes, and the latter two for the francium isotope ^{214}Fr with additional α -decay spectroscopy. Measurements were made using the CRIS technique at the ISOLDE facility in CERN, facilitating some of the most sensitive nuclear structure studies on exotic nuclei to date. Where applicable, experimental results are compared to empirical, and LSSM calculations. The amalgamated data set has presented the opportunity of probing the sturdiness of three doubly-magic nuclei with respect to their local vicinities: ^{56}Ni , ^{78}Ni , and ^{208}Pb .

A magical ^{56}Ni core is confirmed with an electromagnetic-moment analysis of $^{65,67,69}\text{Ga}$, albeit of a soft nature. Comparisons with shell model calculations not only confirm increased mixtures of $\pi 2p_{3/2}/\pi 1f_{5/2}$ configurations and the presence of neutron correlations towards $N = 28$, but they also highlight systematic trends with respect to the neighbouring odd- Z isotope chains. Analyses of neutron-rich species up to ^{82}Ga reveal a rich collection of nuclear-structure effects: a gradual proton-occupation migration from $\pi 2p_{3/2} \rightarrow \pi 1f_{5/2}$ for the odd- A species which culminates in a ground-state spin reversal involving those spins at ^{81}Ga , isomeric presence in ^{80}Ga at $N = 49$, and a kink in the change in the mean-square charge radii for ^{82}Ga at $N = 51$. Overall, this adds further evidence for a robust doubly magic ^{78}Ni core.

Measurements of ^{214}Fr with $t_{1/2} = 5$ ms represents the shortest-lived isotope to have been measured with laser spectroscopy techniques at an online facility, in which the ISOL production mechanism is the limiting factor. With its g factor suggesting a relatively pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration for $I^\pi = 1^-$ and a kink in its charge-radii value at $N = 127$, the magical effects of ^{208}Pb can be observed five protons away.

Beknopte samenvatting

In deze thesis worden de kernspins, de elektromagnetische momenten en de ladingsstraalverschillen van de radioactieve gallium isotopen bestudeerd. Bijkomend worden ook deze laatste twee kerneigenschappen besproken voor ^{214}Fr in combinatie met α -verval spectroscopie. Deze metingen werden uitgevoerd met behulp van de CRIS-techniek in de ISOLDE-faciliteit in CERN, hier vinden vandaag de dag enkele van de meest gevoelige kernstructuur studies plaatst. Waar toepasbaar, worden de experimentele resultaten vergeleken met empirische en LSSM berekeningen. De samengevoegde dataset heeft de mogelijkheid geboden om de robuustheid van de drie dubbel-magische kernen ^{56}Ni , ^{78}Ni en ^{208}Pb in deze omgeving te onderzoeken.

De analyse van de elektromagnetische momenten van $^{65,67,69}\text{Ga}$ duidt op een vrij zachte magische ^{56}Ni kern. Vergelijkingen met schillenmodelberekeningen bevestigen niet enkel een toegenomen menging van $\pi 2p_{3/2}/\pi 1f_{5/2}$ configuraties en de aanwezigheid van neutron correlaties in de richting van $N = 126$, ze geven ook de systematische trends ten opzichte van de naburige oneven- Z -isotopen reeksen aan. Analyse van de neutronrijke isotopen tot en met ^{82}Ga onthullen een rijke collectie aan kernstructuur effecten: een geleidelijke proton migratie van $\pi 2p_{3/2} \rightarrow \pi 1f_{5/2}$ voor de oneven A -isotopen welke zijn hoogtepunt bereikt in een grondtoestandsspin omkering met betrekking tot de kernspinnen rondom ^{81}Ga , isomeren in ^{80}Ga met $N = 49$ en een ladingsstraalkink naar ^{82}Ga met $N = 51$. Algeheel levert dit aanvullend bewijs voor een robuuste magische ^{78}Ni kern.

^{214}Fr ($t_{1/2} = 5$ ms) is het korstlevende isotoop ooit gemeten in een online faciliteit. Hierbij was het ISOL-productiemechanisme de limiterende factor. Met zijn g factor die een relatief pure ($\pi 1h_{9/2} \otimes \nu 2g_{9/2}$) configuratie voor $I^\pi = 1^-$ suggereert en een kink in zijn ladingsstralen bij $N = 127$, kan het magische effect van ^{208}Pb worden waargenomen vijf protonen weg van de schilsluiting.

Contents

Acknowledgements	i
Abstract	v
Beknopte samenvatting	vii
Contents	ix
List of Abbreviations	xiv
List of Figures	xvii
List of Tables	xxiii
1 Introduction	1
2 Theoretical concepts for nuclear structure	7
2.1 Elements of nuclear physics	7
2.1.1 The nuclear shell model	8
2.1.2 Nuclear ground-state properties	13
2.2 Elements of atomic physics	24
2.2.1 Hyperfine structure	25

2.2.2	Isotope and isomeric shift	27
2.3	The concepts of laser spectroscopy	28
2.3.1	Extracting the electromagnetic moments	30
2.3.2	Extracting the changes in the mean-square charge radii	31
2.3.3	Extracting the nuclear spin	33
2.3.4	Summary	35
3	Experimental details for observing hyperfine structures	37
3.1	The ISOLDE facility at CERN	37
3.1.1	Isotope production	39
3.1.2	Isotope ionisation	41
3.1.3	Isotope acceleration and separation	42
3.1.4	Bunched beams with the ISCOOL	42
3.2	Collinear Resonance Ionization Spectroscopy	43
3.2.1	Collinear Laser Spectroscopy	44
3.2.2	Resonance Ionisation Spectroscopy	47
3.3	The CRIS setup at ISOLDE	48
3.3.1	Neutralisation with the charge-exchange cell	49
3.3.2	Laser-ionisation schemes in the interaction region	50
3.3.3	The detection of hyperfine structure	55
3.3.4	Decay-spectroscopy studies at the CRIS setup	56
3.4	Summary	57
4	Analytical tools for the interpretation of hyperfine structure	59
4.1	Summarising CRIS-setup measurements	59
4.1.1	For gallium isotopes	60
4.1.2	For francium isotopes	62
4.2	Treatment of raw data from the CRIS setup	63

4.3	Choice of cost function	65
4.4	Lineshape profiles for laser spectroscopy	66
4.4.1	Voigt lineshapes	67
4.4.2	Crystal Ball lineshapes	68
4.4.3	An asymmetry parameter for lineshapes	73
4.5	Combining multiple measurements of an observable	77
4.6	Summary	78
5	Nuclear magicity near ^{56}Ni	79
5.1	Article I: Probing the $_{31}\text{Ga}$ ground-state properties in the region near $Z = 28$ with high-resolution laser spectroscopy	80
5.2	Characterisations of the reference isotope, ^{71}Ga	101
5.3	Nuclear-spin-assignment analyses for $^{65,67,69}\text{Ga}$	103
5.4	Electromagnetic-moment analyses for $^{65,67,69}\text{Ga}$	106
5.5	Isotope shifts and changes in the mean-square charge radii for $^{65,67,69}\text{Ga}$	110
5.6	Summary	112
6	Nuclear magicity near ^{78}Ni	115
6.1	Nuclear-spin-assignment and electromagnetic-moment analyses for $^{75,79,81}\text{Ga}$	118
6.2	A summary of the hyperfine parameters and electromagnetic moments for $^{75,79,81}\text{Ga}$	121
6.3	Nuclear-spin-assignment and electromagnetic-moment analyses for $^{80,82}\text{Ga}$	124
6.3.1	Paar's parabola rule predictions	125
6.3.2	Two long-lived states in ^{80}Ga	127
6.3.3	The nuclear structure of ^{82}Ga	130
6.4	A summary of the hyperfine parameters and electromagnetic moments for $^{80,82}\text{Ga}$	134

6.5	Isotope shifts and changes in the mean-square charge radii for $^{75,79-82}\text{Ga}$	136
6.6	Summary	138
7	Nuclear magicity near ^{208}Pb	139
7.1	Article II: Laser and decay spectroscopy of the short-lived isotope ^{214}Fr	142
7.1.1	Characterisations of the reference isotope, ^{221}Fr	159
7.1.2	^{214}Fr hyperfine scans	160
7.2	Nuclear-spin-assignment analysis of ^{214}Fr	162
7.2.1	Via Paar's parabola rule	162
7.2.2	Via laser-spectroscopy methods	163
7.2.3	Via decay-spectroscopy methods	165
7.3	g factor of ^{214}Fr	168
7.4	Change in the mean-square charge radii of ^{214}Fr	172
7.5	Summary	175
8	On the path to further understanding nuclear structure	177
8.1	For gallium isotopes	178
8.1.1	$^{61,62}\text{Ga}$: Towards the vicinity of ^{56}Ni	178
8.1.2	$^{83,84}\text{Ga}$: Exploring further past $N = 50$	182
8.1.3	Additional ionisation schemes for performing CRIS on gallium isotopes	186
8.1.4	Future decay spectroscopy of the two long-lived states in ^{80}Ga	194
8.2	For francium isotopes	196
8.2.1	$^{214,218}\text{Fr}$: Characterising the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration	197
8.3	Summary	201

9	Conclusions	203
A	Appendices	205
A.1	Essential quantum-mechanical relations	205
A.2	Derivation of the single-particle Schmidt moments	207
A.3	Compilations of changes in the mean-square charge radii across the nuclear landscape	208
A.4	ν configuration-mixing calculations for the g factor of ^{214g}Fr . .	210
A.5	A spin-assignment-analysis method for hyperfine structures measured with low resolution	218
A.6	A review of francium isotope-shift measurements used for a King- plot analysis	223
A.7	A special case for a pn coupling with $j_p = j_n$	226

List of Abbreviations

AC	Alternating Current.
APIPS	Annular Passivated Implanted Planar Silicon.
BBO	BaB_2O_4 : Beta barium borate.
CCSD	Coupled-Cluster: Singles and Doubles.
CEC	Charge-Exchange Cell.
CERN	Conseil Européen pour la Recherche Nucléaire.
CLS	Collinear Laser Spectroscopy.
COLLAPS	COLlinear LAser SPectroscopy.
CRIS	Collinear Resonance Ionization Spectroscopy.
DCM	4-DiCyanoMethylene -2- methyl-6-(4-dimethyl aminostyryl)-4 <i>H</i> -pyran.
DMSO	DiMethyl SulfOxide.
DSS	Decay Spectroscopy Station.
FC	Faraday Cup.
FWHM	Full Width at Half Maximum.
GPS	General Purpose Separator.
GSI	Gesellschaft für SchwerIonenforschung.
HeNe	Helium-Neon.
HRS	High Resolution Separator.
IDS	Isolde Decay Station.
IKS	Instituut voor Kern- en Stralingsfysica.
IP	Ionisation Potential.
IR	Interaction Region.

ISCOOL	ISOLDE COOLer.
ISOL	I sotope S eparator O n- L ine.
ISOLDE	I sotope S eparator O n- L ine D Evice.
ISOLTRAP	I sotope S eparator O n- L ine T RAP.
LD	L iquid- D rop.
LIST	L aser I on S ource T rap.
LSSM	L arge- S cale S hell- M odel.
MCDF	M ulti C onfiguration D irac- F ock.
MCMC	M onte- C arlo M arkov C hain.
MCP	M ulti- C hannel P late.
MEDICIS	MED ical I sotopes C ollected from ISOLDE .
Nd:YAG	N eodymium- d oped y ttrium a luminium g arner.
NMR	N uclear M agnetic R esonance.
OES	O dd- E ven S taggering.
PIPS	P assivated I mplanted P lanar S ilicon.
PMT	P hoto- M ultiplier T ube.
PSB	P roton S ynchrotron B ooster.
RILIS	R esonance I onization L aser I on S ource.
RIS	R esonance I onisation S pectroscopy.
RMF	R elativistic M ean- F ield.
RMS	R oot M ean S quare.
SHF	S kyrme- H artree- F ock.
SHO	S imple H armonic O scillator.
SPE	S ingle- P article E nergy/ E nergies.
StB	S ignal- t o- B ackground.
TBME	T wo- B ody M atrix E lements.
Ti:Sa	T itanium- S apphire.
UHV	U ltra- H igh V acuum.
UV	U ltra V iolet.
VADLIS	V ersatile A rc D ischarge and L aser I on S ource.
WISArD	W eak I nteraction S tudies with ^{32}Ar D ecay.

List of Figures

1.1	The nuclide chart of observable isotopes according to their primary decay mode.	2
1.2	The colour code used to label hyperfine-spectrum fits throughout this thesis.	5
2.1	Nuclear shell model levels predicted from the simple harmonic oscillator, with the incorporation of the l^2 and spin-orbit interaction terms.	9
2.2	Model spaces used for the: GXPF1, fpg , JUN45, and jj44b interactions.	13
2.3	Allowed hyperfine transitions for the $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transition in ^{71}Ga with $I = 3/2$	29
2.4	Available methodologies for determining the nuclear spin in laser spectroscopy.	33
3.1	A schematic of the current layout of the ISOLDE facility. . . .	38
3.2	A selection of various transition schemes available for laser spectroscopy.	47
3.3	A schematic of the CRIS experimental setup at ISOLDE. . . .	49
3.4	The two-step resonance ionisation scheme used to study neutral francium atoms at the CRIS setup.	51
3.5	The three-step resonance ionisation scheme used to study neutral gallium atoms at the CRIS setup.	53

3.6	Saturation curves for each transition used in the 2015 IS571 gallium experiment at the CRIS setup.	54
3.7	Schematic drawings of the DSS1.2 setup at the CRIS setup. . .	56
4.1	Predicted yields for gallium isotopes measured during the 2015 IS571 experiment.	61
4.2	Typical raw-data formats for wavemeter_ds, cris_ds, and iscool_ds information during a standard CRIS experiment. . .	63
4.3	Normalised Gaussian (dotted line), Lorentzian (dashed line) and Voigt (full line) profiles, each with a FWHM of 25 MHz. . . .	67
4.4	The changes in the shape of a normalised Crystal Ball profile by varying the α , and n asymmetry parameters.	69
4.5	A comparison of fitting with Voigt and Crystal Ball profiles, for selected 2015 IS571 ^{71}Ga measurements.	71
4.6	A comparison of extracted hyperfine observables from fitting selected ^{71}Ga hyperfine-structure measurements from the 2015 IS571 experiment, with Voigt and Crystal Ball profiles.	72
4.7	Selected ^{71}Ga measurements obtained using different 1064-nm pulse energies, from the Litron laser setup during the 2015 IS571 experiment.	72
4.8	Selected ^{71}Ga measurements obtained using different 417.2-nm powers, from the M-Squared SolsTiS setup during the 2015 IS571 experiment.	73
4.9	The effect of the asymmetry parameter a on the lineshape of a Lorentzian distribution.	74
4.10	A comparison of fitting with Crystal Ball, asymmetric-Lorentzian, and Voigt profiles, for selected 2017 IS571 ^{71}Ga measurements.	75
4.11	A comparison of extracted hyperfine observables from fitting selected ^{71}Ga hyperfine-structure measurements from the 2017 IS571 experiment, with Voigt, Crystal Ball, and asymmetric-Lorentzian profiles.	76
4.12	Selected ^{71}Ga measurements obtained using different 417.2-nm powers, from the injection-seeded Ti:Sa laser setup during the 2017 IS571 experiment.	77

5.1	$E(2_1^+)$ and two-nucleon separation energies in the vicinity of the $Z = 28$ and $N = 28$ shell closures.	79
5.2	Temporal trends of extracted ^{71}Ga hyperfine parameters with respect to the start time of the first ^{71}Ga measurement, for each IS571 experiment.	101
5.3	Temporal trends of extracted ^{71}Ga centroid frequencies with respect to the start time of the first ^{71}Ga measurement, for each IS571 experiment.	102
5.4	Hyperfine spectra of $^{65,67,69}\text{Ga}$, alongside A -hyperfine-parameter ratios for $I = 3/2$, $5/2$, and $7/2$ spin assignments.	104
5.5	GXPf1 and JUN45 calculations showing the predicted energies of the lowest-lying $I^\pi = 1/2^-$, $3/2^-$, and $5/2^-$ states for odd- A isotopes between $^{59-71}\text{Ga}$ inclusive.	105
5.6	g factors and electric-quadrupole moments for $^{65,67,69}\text{Ga}$, with additional literature values for the $N = 28 - 40$ region. Comparisons with GXPf1 and JUN45 calculations are also shown.	107
5.7	Available g factors for the $_{29}\text{Cu}$, $_{31}\text{Ga}$, and $_{33}\text{As}$ isotope chains within $N = 28 - 40$, compared with various GXPf1 and JUN45 calculations.	108
5.8	Available electric-quadrupole moments for the isotope-chain pairs: $_{15}\text{Cl}$ and $_{19}\text{K}$, $_{29}\text{Cu}$ and $_{31}\text{Ga}$, and $_{41}\text{Nb}$ and $_{49}\text{In}$, between relevant (sub)shell closures.	109
5.9	Mean-square charge radii for isotopes in the vicinity of the ^{56}Ni region.	111
6.1	$E(2_1^+)$ and two-nucleon separation energies in the vicinity of the $Z = 28$ and $N = 50$ shell closures, near ^{78}Ni	116
6.2	Hyperfine spectra of $^{75,79,81}\text{Ga}$, alongside A -hyperfine parameter ratios and (applicable) g -factors for $I = 3/2$, $5/2$, and $7/2$ spin assignments.	119
6.3	g factors and electric-quadrupole moments of $^{75,79,81}\text{Ga}$ with other odd- A literature values in the $N = 40 - 50$ region. JUN45, jj44b, and fpg calculations are also shown for comparison for both parameters.	122

6.4	Valence-proton distributions and neutron-orbital occupations across the orbitals involved in the JUN45 model space, for the ground states of odd- A $^{59-81}\text{Ga}$	123
6.5	Paar parabolas for selected pn ground-state couplings that are suspected to manifest in ^{80}Ga	125
6.6	Paar parabolas for selected pn ground-state couplings that are suspected to manifest in ^{82}Ga	126
6.7	A hyperfine spectrum of ^{80g}Ga , alongside comparisons of extracted g factors with empirical values for $I = 2 - 7$ spin assignments.	128
6.8	A hyperfine spectrum of $^{80g,m}\text{Ga}$, alongside comparisons involving the extracted A -hyperfine-parameter ratios and subsequent g factors for $I = 2 - 7$ spin assignments.	129
6.9	Hyperfine spectra of ^{82}Ga using ISCOOL collection times of 100 ms and 10 ms.	131
6.10	Goodness-of-fit plots for the ^{82}Ga hyperfine spectra in Figure 6.9 as a function of A_u and B_l , for $I = 1 - 5$ spin assignments. . .	132
6.11	The combined hyperfine spectrum of ^{82}Ga , alongside comparisons of extracted g factors with empirical values for $I = 1 - 5$ spin assignments.	133
6.12	g factors and electric-quadrupole moments of $^{80g,80m,82}\text{Ga}$ with other odd- A literature values in the $N = 40 - 50$ region. JUN45, jj44b, and fpg calculations are also shown for comparison for both parameters.	135
6.13	Mean-square charge radii for isotopes in the vicinity of the ^{78}Ni region.	137
7.1	$E(2_1^+)$ and two-nucleon separation energies in the vicinity of the $Z = 82$ and $N = 126$ shell closures, near ^{208}Pb	139
7.2	A trio of shape coexistence phenomena in the ^{208}Pb region. . .	140
7.3	Changes in the mean-square charge radii for isotopes in the vicinity of the ^{208}Pb region.	141
7.4	Low- and high-resolution hyperfine spectra of ^{221}Fr	159

7.5	Hyperfine spectra of ^{214}Fr , each fitted assuming $I = (1)$ and (2) tentative spin assignments.	161
7.6	Paar parabolas for selected pn ground-state couplings that are suspected to manifest in ^{214}Fr	163
7.7	α -decay spectroscopy of the full atomic and laser-ionised $A = 214$ isotope beam.	165
7.8	Energy-calibration curves for the APIPS and PIPS detectors, used for the collection of α -decay spectroscopy data presented in Figure 7.7.	166
7.9	An overview of the shell-model orbitals which feature past $Z = 82$ and $N = 126$	168
7.10	g factor of ^{214}Fr assuming $I = (1)$ and (2) spin assignments, with other even- A literature values in the $N = 126 - 136$ region. Empirical values for suspected configurations are also shown. . .	171
7.11	Different interpretations of the changes in the mean-square charge radii, for isotope chains immediately before and after $N = 126$	174
8.1	Attempted hyperfine-structure measurements for ^{62}Ga	179
8.2	A simulated spectrum of the hyperfine structure of ^{62}Ga , fitted assuming an $I = 0$ spin assignment.	180
8.3	A simulated spectrum of the hyperfine structure of ^{61}Ga , fitted assuming an $I = 3/2$ spin assignment.	181
8.4	Attempted hyperfine-structure measurements for ^{83}Ga	183
8.5	A simulated spectrum for the hyperfine structure of ^{83}Ga , fitted assuming $I = 3/2, 5/2$, and $7/2$ spin assignments. A corresponding A -hyperfine-parameter ratio comparison is also given.	184
8.6	A simulated spectrum for the hyperfine structure of ^{84}Ga , fitted assuming an $I = 0$ and $I = 3$ ground and isomer state.	185
8.7	Additional transition schemes that could be used for measuring gallium isotopes at the CRIS experiment.	187
8.8	Simulated spectra for the hyperfine structure of ^{71}Ga , for all allowed transitions between $4p\ ^2P_{\frac{3}{2},\frac{1}{2}} \rightarrow 5s, 6s, 7s, 8s\ ^2S_{\frac{1}{2}}$	188

8.9	Simulated spectra for the hyperfine structure of ^{71}Ga , for all allowed transitions between $4p\ ^2P_{\frac{3}{2},\frac{1}{2}} \rightarrow 4s4p^2\ ^4P_{\frac{5}{2},\frac{3}{2},\frac{1}{2}}$	191
8.10	Simulated spectra for the hyperfine structure of ^{80g}Ga , using the $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$ and $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transitions.	192
8.11	Simulated spectra for the hyperfine structure of ^{82}Ga , using the $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$ and $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transitions.	193
8.12	Representative hyperfine-spectrum measurements for the entire structure of ^{80}Ga taken during the 2015 IS571 and 2017 IS571 experiments.	194
8.13	Overlayed hyperfine-spectrum simulations of ^{80g}Ga and ^{80m}Ga , using the $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$, and $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transitions.	195
8.14	Simulated spectra for the hyperfine structure of ^{214m}Fr , using the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition, assuming $I = 1$ and 2 spin assignments for the ground state.	198
8.15	A simulated spectrum for the hyperfine structure of ^{214}Fr , fitted assuming an $I = 1$ and $I = 8$ ground and isomer state.	199
8.16	A simulated spectrum for the hyperfine structure of ^{218}Fr , fitted assuming an $I = 1$ and $I = 8$ ground and isomer state.	200
A.1	Empirical ^{214g}Fr g factors for $I^\pi = (1^-)$ and (2^-) assignments, assuming a maximum of two neutron orbital contributions.	216
A.2	Empirical ^{214g}Fr g -factor relations for $I^\pi = (1^-)$ and (2^-) assignments, assuming a maximum of three neutron-orbital contributions.	218
A.3	A diagram of an example hyperfine spectrum for the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition in francium, applicable for an isotope with nuclear spin of $I \geq 3/2$	219
A.4	A comparison between the original and updated King plots used for the extraction of F and M atomic factors associated with the 422.7-nm transition.	223
A.5	A correlation plot between the gradient (named Order1Coeff) and intercept (named Order1Coeff) of the King plot presented in Figure A.4(b).	224

List of Tables

1	A table containing people whom I wish to thank for making my Ph.D journey so memorable.	ii
2.1	Effective proton and neutron g -factor operators and charges, used for the GXPF1, JUN45, jj44b, and fpg interactions.	12
4.1	A summary of gallium isotope measurements performed at the CRIS setup.	60
4.2	A summary of francium isotope measurements performed at the CRIS setup, relevant for this thesis.	62
5.1	A - and B -hyperfine parameters for ^{71}Ga , representative of weighted means from the amalgamated IS571 data set; the A -hyperfine parameter ratio is also given.	102
5.2	A summary of the uncertainty differences from the A -hyperfine-parameter ratio analysis, and average χ^2_ν values for the full $^{65,67,69}\text{Ga}$ data sets.	105
5.3	Extracted A -hyperfine parameters and magnetic-dipole moments for $^{65,67,69}\text{Ga}$	106
5.4	Extracted B -hyperfine parameters and electric-quadrupole moments for $^{65,67,69}\text{Ga}$	107
5.5	Final isotope shifts and changes in the mean-square charge radii for $^{65,67,69}\text{Ga}$	110

6.1	A summary of the uncertainty differences from the A -hyperfine-parameter ratio analysis, and average χ^2_ν values for the full $^{75,79,81}\text{Ga}$ data sets.	120
6.2	Extracted A -hyperfine parameters and magnetic-dipole moments for $^{75,79,81}\text{Ga}$	121
6.3	Extracted B -hyperfine parameters and electric-quadrupole moments for $^{75,79,81}\text{Ga}$	122
6.4	Extracted A -hyperfine parameters and magnetic-dipole moments for $^{80g,80m,82}\text{Ga}$	134
6.5	Extracted B -hyperfine parameters and electric-quadrupole moments for $^{80g,80m,82}\text{Ga}$	135
6.6	Final isotope shifts and changes in the mean-square charge radii for $^{75,79-82}\text{Ga}$	136
7.1	Extracted A - and B -hyperfine parameters and FWHM's for low- and high-resolution measurements of ^{221}Fr	160
7.2	Extracted A -hyperfine parameters and isotope shifts for ^{214}Fr	161
7.3	Extracted intensity ratios between the two hyperfine resonances observed in the ^{214}Fr spectra displayed in Figure 7.5.	164
7.4	Representative single-particle magnetic-dipole moments for the shell-model orbitals featuring in the extended model space from Figure 7.9.	169
7.5	Extracted g factors for ^{214g}Fr assuming tentative $I = (1)$ and (2) spin assignments, compared with LSSM calculations and empirical values for various pn configurations.	170
7.6	Final changes in the mean-square charge radii for ^{214}Fr , assuming the tentative $I = (1)$ and (2) spin assignments.	172
7.7	Additional data for changes in the mean-square charge radii immediately before and after $N = 126$	173
7.8	Available ξ_{even} and ξ_{odd} parameters for elements in the vicinity of $N = 126$	173
8.1	Expected $^{61,62}\text{Ga}$ yields using ZrO_2 target material, alongside background-limit estimates.	179

8.2	Expected $^{83,84}\text{Ga}$ yields using UC_x target material, alongside background-limit estimates.	183
8.3	Expected $^{214g,218g}\text{Fr}$ yields using UC_x target material, alongside background-limit estimates for isobaric radium.	198
A.1	References which contain $\delta\langle r^2 \rangle$ for the isotope chains featuring in Figures 5.9, 6.13, and 7.3.	209
A.2	Extracted F_{422} and M_{422} factors resulting from the progressive addition of updated data and new analysis techniques.	224
A.3	$\delta\langle r^2 \rangle$ comparisons between previously-published and newly re-calculated CRIS values, with TRIUMF values.	225

Chapter 1

Introduction

Unbeknownst to more people than one may suspect, the very definition of nuclear physics is completely different to its usual connotation with atomic bombs and nuclear power stations: it studies matter down to the sub-atomic level and beyond, examining not only the constituent nuclei but also their interactions. Its beginnings originate from attempting to comprehensively describe this atomic picture with Ernest Rutherford more than 100 years ago [Rut14]. Originally imagined as positively charged centres with orbiting negatively charged electrons, there have since been huge strides in the refining of the atomic description and understanding of nuclear structure phenomena. It is important to state that with respect to an atomic system, one is not just confined to within the realms of elements in a periodic table. Each known element can possess a variable number of neutrons, with each individual flavour known as an *isotope*. The playground of such systems available for study is therefore vast; the nuclide chart given in Figure 1.1 represents at least 3000 species [Sót19] and subsequently, variations in the structure of the atomic nucleus.

One of the biggest continuing questions in nuclear physics is how the structure of the nucleus manifests with different quantities of nucleons. Arguably the most celebrated description of nuclei comes from the pioneering work of M. Goeppert-Mayer¹: the nuclear-shell model. It predicts an enhanced configurational stability for nuclei with a *magic* number of protons and/or neutrons: 2, 8, 20, 28, 50, 82, and 126. Primarily inferred from the studies of stable and long-lived isotopes, recent technological advancements and developments into radioactive

¹Alongside E. P. Wigner and J. Hans D. Jensen, the nuclear shell-model theory won the Nobel Prize for Physics in 1963 [Nob18].

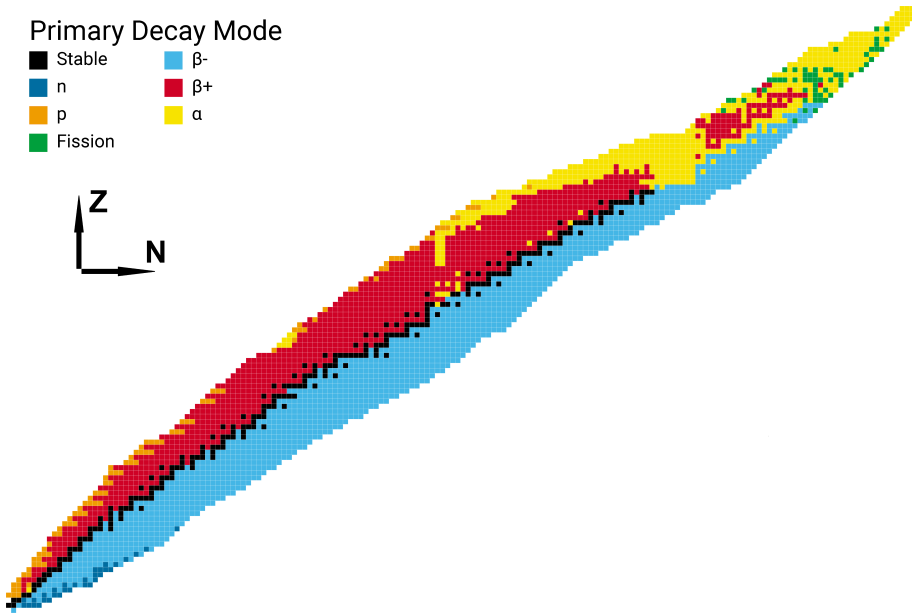


Figure 1.1: The nuclide chart of isotopes according to their primary decay mode. Data is taken from the most recent Atomic Mass Evaluation in Ref. [Aud17] and includes both observations and predictions. Figure adapted from Ref. [Sim17]².

beam facilities over the years have helped to cement their magical reputation across the nuclide chart.

Furthermore, the property of magicity itself can prove to be a dynamic property [Dob94; Sor08], particularly in regions where isotopes possess large N/Z ratios and unprecedented nuclear structures. Generally, such manifestations can be explained by the actions of some nucleon-nucleon tensor monopole interaction [Ots01; Ots05; Ots10]. The $N = 8$ shell gap for example, disappears in ^{12}Be because of a less-bound $\nu 1p_{1/2}$ shell as p -type protons are depleted from the nuclear system [Nav00]. A similar manifestation has also been observed for $N = 28$ in ^{42}Si , due to the compression of the proton and neutron levels from the attractive and repulsive interactions between the $\pi 1d_{5/2}$ and neutron- pf shells [Bas07]. Impromptu occurrences are also just as intriguing, such as the appearance of an $N = 16$ shell gap [Oza00] in $^{23,24}\text{O}$ due to a large energy gap

²Aesthetically pleasing interactive versions can be found at The Colourful Nuclide Chart: <http://people.physics.anu.edu.au/~ecs103/chart/>, created by Edward Simpson at The Australian National University. They can be created with respect to a wide range of fundamental nuclear structure properties.

between the $\nu 2s_{1/2}$ orbit with the $\nu 1d_{5/2}$ and $\nu 1d_{3/2}$ doublet [Cor04; Kan09; Hof09]. The $N = 32$ subshell gap has recently drawn attention; along with effects from three-nucleon ($3N$) forces, it stems from an attractive $\pi 1f_{7/2}$ - $\nu 1f_{5/2}$ proton-neutron (pn) interaction which weakens with the depletion of the $\pi 1f_{7/2}$ orbital [Ste13; Hag16]. A multitude of studies has (so far) been dedicated to characterise its magnitude for several isotones ranging from $_{18}\text{Ar}$ to $_{24}\text{Cr}$, in which discontinuities have been seen in the: $E(2_1^+)$ energies [Pri01; Jan02; Ste15], mass measurements [Gal12; Wie13; Xu19], charge radii [Gar16], and $B(E2; 0_1^+ \rightarrow 2_1^+)$ reduced-transition probabilities [Din05; Bür05]. $N = 34$ is also postulated to be a subshell gap based on similar theoretical principles, however at present this has only been observed for $_{18}\text{Ar}$ and $_{20}\text{Ca}$ with respect to: $E(2_1^+)$ energies [Ste13; Liu19], and mass measurements [Wie13; Mic18]. Other questions such as: the appearance of other non-standard magic numbers in heavier mass regimes [Sor08], as well as what comes after $Z = 82$ and $N = 126$ [Ben99; Hee02; Ćwi05; Sor08] have yet to be answered. This reinforces the golden pinnacle of scientific endeavour: the continuous and thorough scrutiny of any given proposal or statement.

Special attention is reserved for nuclei which possess magic numbers for both N and Z , commonly referred to as *doubly-magic*. Suspected systems not only exhibit exceptional stability, but they can also be modelled as spherical closed shell configurations. While some such as $_{20}^{40}\text{Ca}$, $_{28}^{56}\text{Ni}$, and $_{82}^{208}\text{Pb}$ have been extensively studied throughout the years, investigations on others like $_{28}^{78}\text{Ni}$ have only started very recently. A variety of experimental techniques are available for obtaining the nuclear properties mentioned above, which are required to investigate the phenomena of shell evolution. Nuclear masses, arguably the most fundamental characterisation of the nucleus, can be measured at mass-spectrometry setups (see Ref. [Aud17]). Other properties like excited states and transition probabilities, representing the finer nuances of nucleon interactions, are obtained from transfer-reaction and decay-spectroscopy setups. Laser-spectroscopy techniques yield parameters such as the electromagnetic moments, charge radii, and ground-state spins, helping to characterise the nucleon configuration and shape of the nucleus as a whole. Its nuclear-model independence is one of its most elegant features, thus allowing for a thorough, independent investigation into not only the magical sturdiness of doubly-magic systems, but also how tenacious their magical properties are with respect to each of their local vicinities.

Following this introduction, chapter two sets out the theoretical framework with respect to nuclear physics, atomic physics, and laser spectroscopy. The experimental technique used to probe nuclear structure phenomena is introduced in chapter three, followed by a discussion of the tools used for the analysis of subsequent data in chapter four. Three doubly-magic shell closures are then

explored: ^{56}Ni , ^{78}Ni , and ^{208}Pb , in chapters five, six, and seven, respectively. Individual motivations and statuses regarding their magical characterisations are also included in these chapters. Results from data collected on gallium (first two) and francium (third) isotopes will bring fresh discussions and insights into the current understanding regarding the magical character of the aforementioned doubly-magic nuclei. Finally, chapters eight and nine serve as the concluding remarks: first with an outlook of possible new nuclear structure that can be obtained with current capabilities, and then a summary of the findings and discussions presented in this thesis.

The following peer-reviewed articles mentioned below have been included into this thesis, in order to show the contributions already made in its relevant area of nuclear structure. Unlike their original double-column format as dictated by the journal, they are displayed in a single-column format for easier reading:

I Laser and decay spectroscopy of the short-lived isotope ^{214}Fr in the vicinity of the $N = 126$ shell closure.

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.
Physical Review C, **94**, 054305 (2016).

II Probing the $_{31}\text{Ga}$ ground-state properties in the region near $Z = 28$ with high-resolution laser spectroscopy.

G.J. Farooq-Smith, A.R. Vernon, J. Billowes, C.L. Binnersley, M.L. Bissell, T.E. Cocolios, T. Day Goodacre, R.P. de Groote, K.T. Flanagan, S. Franchoo, R.F. Garcia Ruiz, W. Gins, K.M. Lynch, B.A. Marsh, G. Neyens, S. Rothe, H.H. Stroke, S.G. Wilkins, and X.F. Yang.
Physical Review C, **96**, 044324 (2017).

While these articles are written in American English, the rest of this thesis is (mostly) written in British English and explains any grammatical and spelling differences.



Figure 1.2: The colour code used to label hyperfine-spectrum fits throughout this thesis.

N.B.: Differences between results featuring in a published article and those displayed in the in-depth analysis presented afterwards may occur, due to one (or both) of the following explanations:

- Improved analysis codes for data handling, summing, and analysis, throughout the research period.
- Additional data for a particular isotope measurement only available after the publication of a research article.

Ultimately, results featured in this thesis are consistent with those already published and do not significantly change the physics discussions already made. Excluding the articles mentioned above, the fitting of real and simulated hyperfine-structure spectra adheres to a spin-dependent colour code as defined in Figure 1.2; where appropriate, the reader will be reminded of these choices.

Clarifications: Compared to the printed copies available within the IKS department at KU Leuven, the following mistakes have been addressed:

- p. 122: Added the missing x -axis tick labels, in Figure 6.3(a).
- p. 191: Replaced $4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$ with $4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$, in the caption of Figure 8.9.
- p. 198: Replaced ^{221}Ga with ^{221}Fr , in the x -axis labels of Figure 8.14.
- p. 200: Replaced ^{214g}Fr with ^{218g}Fr , in the x -axis label of Figure 8.16.
- p. 216: Replaced $\nu 1g_{9/2}$ with $\nu 2g_{9/2}$, in the x -axis labels of Figure A.1.
- p. 218: Replaced $\nu 0i_{11/2}$ with $\nu 1i_{11/2}$, in the caption of Figure A.2.

Chapter 2

Theoretical concepts for nuclear structure

In a nutshell, the essence of laser spectroscopy is to examine isotopes through their atomic levels, with sufficient sensitivity for probing the behaviour of the constituent protons and neutrons and their interactions. These valuable insights are crucial input for the (often-imagined) symbiotic relationship between experimental and theoretical nuclear-physics realms, essential for explaining some of the most complex and challenging problems. This first main chapter will deal with the nuclear and atomic theory that are pivotal for the interpretations made later in this thesis.

2.1 Elements of nuclear physics

Many real-world applications rely on an intimate understanding of nuclear-physics concepts: from providing energy with nuclear power, to cancer treatments in hospitals with nuclear medicine, to radionuclide dating for geological and archaeological purposes. Essentially, nuclear physics studies the behaviours and interactions that occur between the constituent nucleons for a particular nucleus. Some models and theoretical concepts for this branch of physics are given in the next couple of sections.

2.1.1 The nuclear shell model

Structural descriptions of nuclei can be inherently complicated due to the number of constituent single particles to account for, effectively resulting in a mesoscopic many-body problem. Although an increasingly daunting task for heavy nuclei, the observation of patterns in many experimental observables provides a starting-point for a theoretical framework. As introduced in Chapter 1, it has long been known that nuclei with a magic number of protons and/or neutrons: have larger-than-average cosmic abundances relative to isotopes residing nearby (see Refs. [Seu56; Bur57]), are more tightly bound nuclei¹, or have higher first excited 2^+ state energies (applicable to even- N, Z nuclei only). Discontinuities in these observables are the main indication for nuclear shell closures. This led to the development of the *nuclear shell model*: a quantum mechanical description in which the single particles sequentially occupy shells each separated by an energy gap, while obeying the Pauli exclusion principle. When a particular shell is completely filled and the energy gap to the next available shell is sufficiently large, the resulting nucleus can be considered to be ‘magic’ and possess an enhanced stability with respect to its nearest neighbours.

Akin to all quantum mechanics problems, defining the nuclear shell model requires knowledge of its Hamiltonian, H . Generally, its total can be concisely described by the sum of the kinetic T and potential V energies of all the constituent nucleons:

$$H |\Psi\rangle = (T + V) |\Psi\rangle = E |\Psi\rangle. \quad (2.1)$$

The latter V term encapsulates all possible inter-nucleon interactions within the nucleus. Even if limited to just including two-body interactions, it is still notoriously challenging to evaluate. Moreover, it is known that three-body forces should also be included in order to successfully describe the structure of the nucleus [Ham13]. However the introduction of a mean-field potential U in (2.1) yields:

$$\begin{aligned} H &= \left[T + \sum_i U_i \right] + \left[V - \sum_i U_i \right] \\ &= H_0 + H_{res}. \end{aligned} \quad (2.2)$$

¹Differences between one- or two-proton/-neutron separation energies, $S_{1p/1n}$ and $S_{2p/2n}$ respectively, can be used to examine the changes across isotone and isotope chains. In particular, it is intuitive to look at the changes between two mass units in order to isolate nucleon pairing effects.

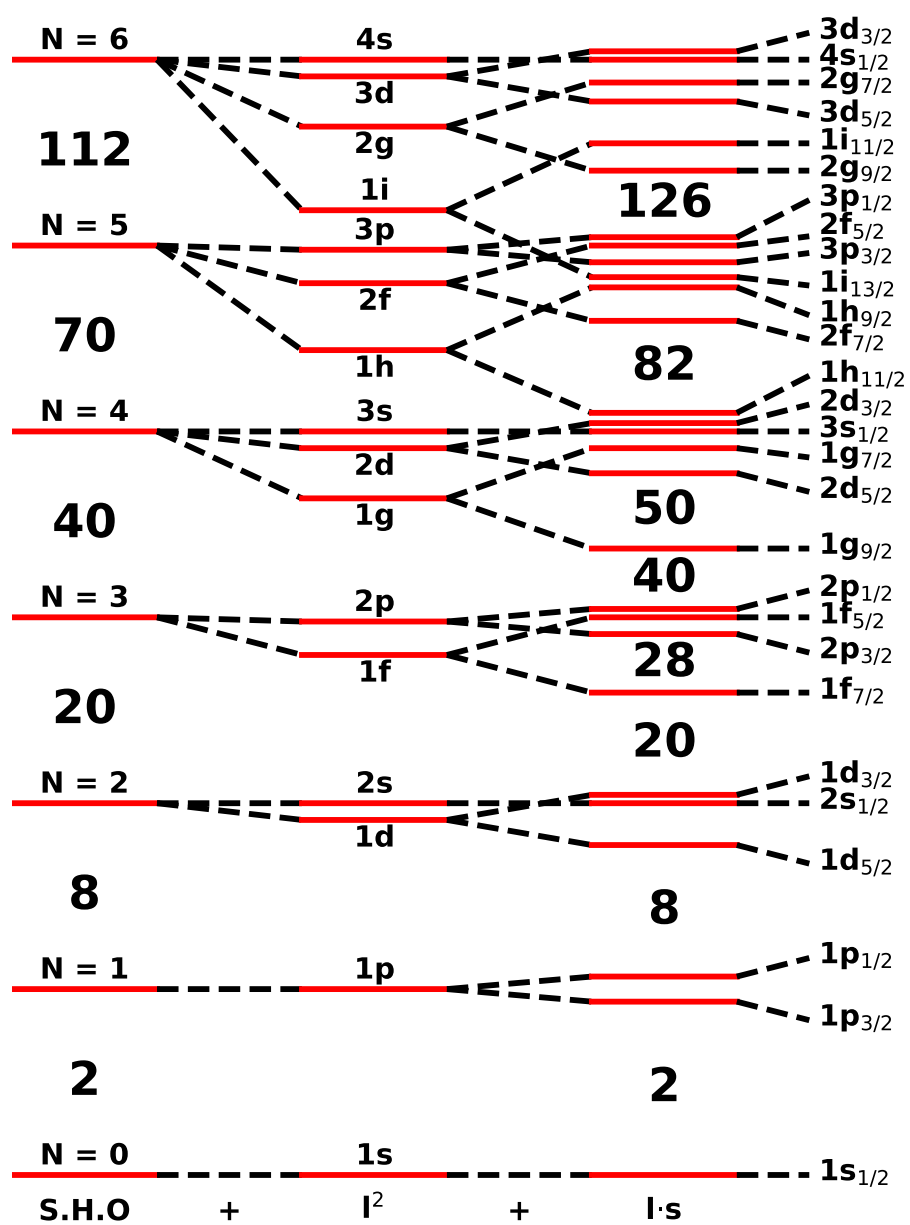


Figure 2.1: Nuclear shell model levels predicted from the simple harmonic oscillator, with the incorporation of the l^2 and spin-orbit interaction terms. See text for further details.

This inclusion has powerful ramifications: H can now be described as the total independent behaviour of each nucleon acting under a potential created by all the others, with the residual effects acting as an additive perturbation to the system.

A long-standing simplification of (2.2) neglects any perturbation effects and is known as *the independent-particle shell model*. Dealing with just $H = H_0$, only a well-suited potential U needs to be chosen and of course within physics, one cannot ignore the temptation of the Simple-Harmonic Oscillator (SHO) potential:

$$U(r) = \frac{1}{2}m\omega^2 r^2, \quad (2.3)$$

resulting in the construction of the left-most level scheme in Figure 2.1. While this reproduces the magic numbers observed in light systems, heavier magic numbers such as 28 and 50 are not successfully generated. Further modifications are needed for (2.3) [May49]:

$$U(r) = \frac{1}{2}m\omega^2 r^2 + Cl^2 + D\mathbf{l} \cdot \mathbf{s}, \quad (2.4)$$

which take the forms of an l^2 term and an $l \cdot s$ spin-orbit term. Both terms introduce a degree of non-degeneracy to the states: the first splits harmonic oscillator levels according to all possible l values, while the coupling of the nucleon spin angular momenta $s = \pm 1/2$ in the second splits the level into a doublet state, such that nucleons with $j = l + 1/2$ have a lower energy than $j = l - 1/2$. This progression² is also documented in Figure 2.1 and ultimately reproduces the familiar set of magic numbers which can be categorised into two types³: those borne from the simple harmonic oscillator (2,8,20), and those which require the spin-orbit interaction (28,50,82,126). Systems with these numbers of nucleons + one-particle/-hole are some of the most effective probes for their suspected magicity, because their nuclear structures and ground-state properties should be wholly determined by the properties of this valence particle/hole; generally this is indeed the case. However for isotopes which are

²Greater energy deviations with the l^2 term and stronger splittings between spin-orbit levels are seen with increasingly larger l levels.

³Although not strictly a shell closure, magical-like behaviour can be seen in the trends of ground-state properties at $N = 40$ and is regarded as a sub-shell closure. See Chapter 5 for further details.

situated well within the confines of magic shell closures, or describing the more intricate effects of nuclear structure, this methodology alone is not sufficient.

Another (necessary) approach is by using *the interacting shell model* in which the H_{res} terms are included in the evaluation of (2.2). Although this description requires the inclusion of all possible nucleon-nucleon interactions within the nucleus, this poses an impossible task for increasingly heavy systems with their exponentially-expensive computational times. It is usually conventional to exclude certain orbits which are suspected to not contribute to the nuclear structure: a closed core of fully occupied orbitals (usually doubly-magic systems), and an upper space of orbitals too far away in energy to interact. The remaining orbitals are known as the *model space* for a certain interaction, in which the valence particles are free to reside anywhere to make a configuration ϕ_i . Thus, the total wavefunction calculated for a given isotope is defined as:

$$\Psi = \sum c_i \phi_i, \quad (2.5)$$

where c_i represents the probability for the corresponding configuration. While descriptions of nuclei close to shell closures are expected to be defined by one relatively-pure configuration, those situated in mid-shell regions likely possess several equally-dominating configurations, more widely referred to as configuration mixing.

The simplification of confining interactions to within a model space however, can profoundly affect the calculated electromagnetic moments, in which deformation and core-excitation effects may not be completely accounted for. Effective charges (q_π and q_ν), and effective spin ($g_{s,eff}$) and orbital g -factors ($g_{l,eff}^\pi$ and $g_{l,eff}^\nu$), both for the proton and neutron, are thus usually introduced to compensate for these effects for the magnetic-dipole and electric-quadrupole moments respectively. This essentially involves a numerical deviation from the free-proton and -neutron charges and g -factors: $q_\pi = 1.0$, $q_\nu = 0.0$, $g_{s,eff} = 1.0g_{s,free}$, $g_{l,eff}^\pi = 1.0$, and $g_{l,eff}^\nu = 0$, which hypothetically should be used for increasingly smaller valence spaces. The choice of effective values are dependent on the shell-model interaction used; values for several Large-Scale Shell-Model (LSSM) interactions are given in Table 2.1.

Large-scale shell model interactions

A variety of LSSM interactions that can be used to help interpret the nuclear structure of isotopes is displayed in Figure 2.2. Specifically, they have all been selected to be used in-conjunction with the study of gallium isotopes: GXPF1

and JUN45 for the neutron-deficient ($N \leq 40$) isotopes, and JUN45, jj44b, and *fp*g for the neutron-rich ($N \geq 40$) species. Their comparisons with experimental data will be shown later in Chapters 5.4, 6.2, and 6.4. It is remarked that only the GXPF1 and JUN45 interactions are readily available on the IKS cluster.

The GXPF1 interaction [Hon02a] is primarily used for comparison of nuclear data in the $A \sim 40 - 71$ mass region. Assuming a ^{40}Ca core, this interaction is represented by four Single-Particle Energies (SPE) from the *pf* shell: $1f_{7/2}$, $2p_{3/2}$, $1f_{5/2}$, and $2p_{1/2}$, along with 195 Two-Body Matrix Elements (TBME) whose starting parameters are derived from a realistic interaction based on the Bonn-C potential [Hjo95]. Using a fitting procedure that is defined in the aforementioned Ref. [Hon02b], 70 selected linear combinations of SPE and TBME are fitted to ~ 700 binding and excitation energies of 87 nuclei in the $^{47}\text{Ca} - ^{65}\text{Ge}$ region. It has already demonstrated successful comparisons with a variety of fundamental nuclear properties such as the electromagnetic moments, binding energies, and $E(2_1^+)$, however these agreements become limited as the end of the *pf* shell is approached. For lighter-mass nuclei typically with $Z \leq 28$, an analogue version known as GXPF1A is more commonly used; further details can be found in Ref. [Hon05]. Meanwhile, the JUN45 interaction [Hon09] is more appropriate for a heavier mass region of $A \sim 60 - 100$. Starting with ^{56}Ni as its core, it also possesses four SPE in its model space, but from: $2p_{3/2}$, $1f_{5/2}$, $2p_{1/2}$, and $1g_{9/2}$. The same re-normalised Bonn-C potential described above is used to derive the starting parameters, but for 133 TBME which are coupled with an $(A/58)^{-0.3}$ mass dependence. A single-valued decomposition method is used to fit selected $l = 45$ linear combinations of SPE and TBME to $e = \sim 400$ experimental energies for $n = 69$ nuclei with $Z = 28 - 30$, $N = 28 - 50$ and $Z = 28 - 50$, $N = 48 - 50$ (overall $A = 63 - 96$). The overall Root-Mean Square (RMS) deviation of this fit is ~ 185 keV.

While the jj44b and *fp*g interactions cannot be accessed by the IKS cluster, they are also included in this thesis for completeness. The jj44b interaction [Bro10] uses the same model and interaction spaces as for JUN45, along with the same methodology already described above. However with: $l = 30$, $e = \sim 600$, and $n = 77$, its RMS deviation is ~ 250 keV. Although unpublished, it has been widely implemented in several experimental works [Che10b; Che10a;

Table 2.1: Effective proton and neutron g -factor operators and charges, used for the GXPF1, JUN45, jj44b, and *fp*g interactions.

Interactions	g_s ($g_{s,free}$)	$g_l(p)$	$g_l(n)$	q_π	q_ν
GXPF1	0.9	1.1	-0.1	1.5	0.5
JUN45; jj44b; <i>fp</i> g	0.7	1.0	0.0	1.5	1.1

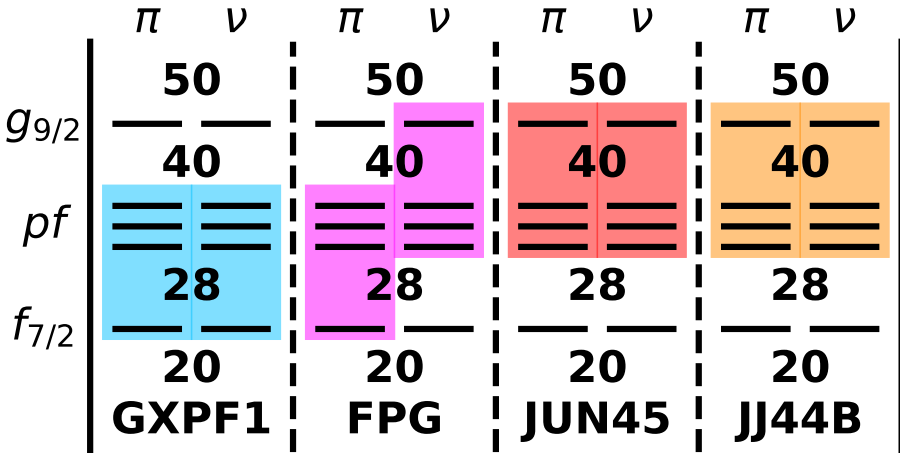


Figure 2.2: Model spaces used for the: GXPF1 (blue), fpg (magenta), JUN45 (red), and jj44b (orange) interactions. See text for their further details.

Man11a; Pro12; Muk17]. The fpg interaction [Sor02] uses a similar model and interaction space as for GXPF1, however with all eight neutrons residing in the $\nu 1f_{7/2}$ orbital frozen (thus, using a ^{48}Ca core) and the addition of the $\nu 1g_{9/2}$ shell. Further information can be found in Refs. [Sor02; Sri11]; the latter aforementioned reference in particular, has performed such calculations for several gallium isotopes and has intended to expand on the existing calculations in Refs. [Che10b; Che10a; Man11b]. Relevant comparisons with experimental data collected at the CRIS setup will be shown later in Chapter 6.2.

For the heavier isotopes like francium, there are no readily available interactions for the description of their fundamental properties and thus, theoretical studies are referenced where appropriate for further comparison. As already mentioned in Chapter 2.1.1, the complexities of the many-body problem become more apparent with heavier nuclei; recently however, shell-model calculations involving a large model space have successfully calculated the electromagnetic moments of neutron-deficient $_{80}\text{Hg}$ isotopes [Mar18a; Sel19].

2.1.2 Nuclear ground-state properties

While the many-body picture for most accessible isotopes is a complex problem to solve, over the years there has been significant progress in quantifying their fundamental nuclear properties. This next section will give an overview of these observables.

The nuclear spin

Following the shell-model description given in Chapter 2.1.1, each of the i single-particle states that constitutes a given nucleus is characterised with a total angular momentum j_i and parity π_i :

$$j_i = l_i + s_i \quad \text{and} \quad \pi_i = (-1)^{l_i}, \quad (2.6)$$

both of which depend on the orbital l_i and spin s_i angular momenta of the orbital in which a particular nucleon resides. Discerning the nuclear spin and parity of a nuclear state, normally expressed together as I^π , is done by including all i nucleons: taking a vector sum for all j_i , and a multiplication for all π_i . While this may seem cumbersome, especially for the heavier nuclear systems, nucleons have a strong tendency to form pairs by virtue of the pairing interaction. They couple to $I^\pi = 0^+$ states with zero angular momentum, ultimately meaning that the core nucleons can be assumed to sum up to a total spin of zero. Thus, only the valence nucleons need to be considered, which leads to the following two generalities:

- All even- N , even- Z nuclei have a ground-state spin and parity of $I^\pi = 0^+$.
- The ground-state spin and parity of odd- A isotopes are both dependent on the shell-model orbital(s) occupied by the unpaired valence nucleon(s), as well as its/their seniority.

The situation becomes more involved for odd- N , odd- Z nuclei. Its ground-state I value can possess any value from the vector coupling of the individual proton and neutron spins j_π and j_ν , while its parity π is the product of the two parities π_{j_π} and π_{j_ν} :

$$I = |j_\pi - j_\nu| \quad \text{to} \quad j_\pi + j_\nu, \quad \text{and} \quad \pi = \pi_{j_\pi} \otimes \pi_{j_\nu}. \quad (2.7)$$

Whichever candidate in (2.7) lies lowest in energy, is the one that forms the ground state for the odd-odd nucleus concerned. One methodology commonly utilised to qualitatively predict the likeliest ground-state spin and parity for such nuclei is Paar's parabola rule. Although detailed in Ref. [Paa79], the following passage aims to provide a summarised and condensed version of the derivation for the purpose of brevity.

In general for a given $(\pi j_p \otimes \nu j_n)$ proton-neutron (pn) valence coupling, the energy differences between the allowed total- I spin states calculated from (2.7) follow a parabolic trend. This originates from phonon exchanges between the valence nucleons, in particular from spin-vibrational 1^+ and quadrupole 2^+ contributions. With energy differences of δE_1 and δE_2 respectively, the total energy perturbation δE for a particular I spin state is given by their sum:

$$\begin{aligned} \delta E &= \delta E_{1+} & + \delta E_{2+} \\ &= \frac{\alpha_{1+}}{4} \left[\frac{(1-\xi)I(I+1)-C}{(j_p+1)(j_n+1)} \right] + \frac{\alpha_{2+}\nu}{4} \left[\frac{1}{3} - \frac{C^2+C}{4j_p(j_p+1)j_n(j_n+1)} \right], \end{aligned} \quad (2.8)$$

where α_{1+} and α_{2+} represent corresponding interaction strengths, approximated as:

$$\begin{aligned} \alpha_{1+} &\approx \frac{160}{A} U_{j_p} V_{j_p} U_{j_n} V_{j_n}, \\ \alpha_{2+} &\approx 4|(U_{j_p}^2 - V_{j_p}^2)(U_{j_n}^2 - V_{j_n}^2)|, \end{aligned} \quad (2.9)$$

with $U_{j_{p,n}}^2$ and $V_{j_{p,n}}^2 = 1 - U_{j_{p,n}}^2$ quantifying the emptiness and fullness factors⁴ of the proton and neutron orbitals involved in the pn coupling, for an isotope with mass A . In addition for (2.8): $C = I(I+1) - j_p(j_p+1) - j_n(j_n+1)$, ξ is defined by:

$$\xi = \begin{cases} \frac{(j_p+1)(j_n+1)}{j_p j_n}, & \text{if } \mathcal{N} = +1 \\ 1, & \text{if } \mathcal{N} = -1 \\ -\frac{j_n+1}{j_n}, & \text{if } \mathcal{N} = 0; j_p - l_p = -1/2 \\ -\frac{j_p+1}{j_p}, & \text{if } \mathcal{N} = 0; j_p - l_p = +1/2, \end{cases} \quad (2.10)$$

which depends on the Nordheim number $\mathcal{N} = j_p + j_n - l_p - l_n$ for the coupling, and ν is the occupation number. Dependent on the natures of j_p and j_n , this final parameter is just equivalent to a sign convention:

- For particle-particle and hole-hole configurations: $\nu = +1$.

⁴Taking ^{214}Fr as an example with 5 protons in the $\pi 1h_{9/2}$ orbital and 1 neutron in the $\nu 2g_{9/2}$ orbital: $U_{j_p}^2 = 5/10$, $V_{j_p}^2 = 5/10$, $U_{j_n}^2 = 1/10$, and $V_{j_n}^2 = 9/10$.

- For all other particle-hole configurations: $\nu = -1$.

It is usually common, if not prudent, to include all configurations that are suspected to tangibly contribute to the total nuclear wavefunction, in a Paar parabola analysis. Any instances of alike spin assignments originating from different pn couplings will have a tendency to repel each other in energy space and can push hitherto unsuspected states towards the ground state. The occurrence of two or more nuclear states laying low in energy may also infer the presence of isomerism. Although cursory, this methodology can be used as a starting point for any logic-based arguments with respect to any experimental observations; applications for the isotopes $^{80,82}\text{Ga}$ and ^{214}Fr can be found in Chapters 6.3.1 and 7.2.1, respectively.

The magnetic-dipole moment and the g factor

The orbital magnetic-dipole moment of an orbiting charged particle is classically related to the orbital angular momentum along the z -direction $\mathbf{l}_z$:

$$\boldsymbol{\mu} = \frac{e\mathbf{l}_z}{2m}, \quad (2.11)$$

in which the expectation value of $\mathbf{l}_z$ is defined as $m_L\hbar$. Additionally, e and m define the charge and mass of the particle respectively. It is usually customary to express this quantity in terms of the nuclear magneton μ_N ⁵ with $m = m_\pi$ as the mass of the proton in (2.11). Quantum mechanically, this description can be extended by considering the intrinsic spin s of the particle. For an ensemble of $Z + N = A$ nucleons, the magnetic-dipole operator $\boldsymbol{\mu}$ is expressed as:

$$\boldsymbol{\mu} = \left(\sum_{i=1}^A g_L^i \mathbf{L}^i + \sum_{i=1}^A g_S^i \mathbf{S}^i \right) \mu_N, \quad (2.12)$$

where g_L and g_S represent the orbital and spin gyromagnetic (g) factors, and are different for the two flavours of nucleons i . For the orbital L values: $g_L^\pi = 1$ and $g_L^\nu = 0$, which are related to the different net charge properties for protons and neutrons. Concerning spin S types: $g_S^\pi = +5.585694702(17)$ and $g_S^\nu =$

⁵Two types exist: the nuclear magneton $\mu_N = \frac{e\hbar}{2m_\pi} = 5.050783699(31) \times 10^{-27}$ J/T, and the Bohr magneton $\mu_B = \frac{e\hbar}{2m_e} = 9.274009994(57) \times 10^{-24}$ J/T.

$-3.82608545(90)$, evaluated for free nucleons in which L cannot contribute. It is also trivial to deduce the relationship between μ and the g factor from (2.12):

$$\mu = gI\mu_N. \quad (2.13)$$

Alternatively for (2.12), the magnetic-dipole moment of a nucleus with spin I can be obtained from the expectation value of the z -component with the projection $M = I$:

$$\begin{aligned} \mu &= \langle I, M = I | \boldsymbol{\mu} | I, M = I \rangle \\ &= \sqrt{\frac{I}{(I+1)(2I+1)}} \langle I || \boldsymbol{\mu} || I \rangle, \end{aligned} \quad (2.14)$$

where obtaining the final expression involves the application of the Wigner-Eckhart theorem, the details of which can be found in Appendix A.1. The I -dependence of (2.14)⁶ means that nuclear states with $I = 0$ do not possess a magnetic-dipole moment; the same also applies to paired nucleons which have a strong tendency to couple to $I = 0$ (with equal and opposite values of m_l). For n particles within a particular single-particle orbital j :

$$\frac{\mu(j^n, I)}{I} = \frac{\mu(j)}{j}, \quad (2.15)$$

thus meaning that the g factor should remain invariant with respect to the value for just a single particle in that orbital. Staying within the realms of single-particle behaviour, the expressions:

$$\begin{aligned} \mu(l+1/2) &= \left[\left(j - \frac{1}{2} \right) g_l + \frac{1}{2} g_s \right], \\ \mu(l-1/2) &= \frac{j}{j+1} \left[\left(j + \frac{3}{2} \right) g_l - \frac{1}{2} g_s \right], \end{aligned} \quad (2.16)$$

⁶Different theoretical conventions exist regarding the relationship between the Wigner-Eckhart theorem and reduced matrix elements, usually involving a factor of $\sqrt{2I+1}$ in the denominator.

can be deduced from (2.14) (see Appendix A.2 for further details) to estimate the single-particle magnetic-dipole moments and are known as the *Schmidt moments* [Ari54]. In reality however, many isotopes do not possess single-particle characteristics and thus, their g factors deviate significantly from this description [Miy51; Flo52; Bli54]; this can be for two reasons. The first is attributed to additional effects originating from the nuclear medium which are neglected from the Schmidt-estimate descriptions in (2.16) to the total magnetic-dipole moment, such as meson-exchange currents and core-polarisation effects for example [Bro87; Cas90]. Effective g -factor operators are thus introduced for simplicity, rather than employing a modified magnetic-dipole moment operator in (2.12) which would otherwise complicate magnetic-dipole moment calculations. These effective values, in essence, encapsulate the aforementioned nuclear effects; used in (2.14) results in *effective Schmidt moments*, or effective single-particle estimates. Typical values used are: $g_{s,eff} \approx 0.6 - 0.9g_{s,free}$, and $g_{l,eff}^\pi \approx 1.1$, $g_{l,eff}^\nu \approx -0.1$. Those used in shell-model calculations described in Chapter 2.1.1 can be found in Table 2.1. A second explanation is the incorrect assumption of describing a nuclear state with a pure single-particle wavefunction; it can be entirely possible for others to contribute towards the total magnetic-dipole moment, giving rise to configuration mixing. These effects however, are expected to be minimal for isotopes residing on or near proton-/neutron-shell closures, which should exhibit single-particle behaviour. Overall, this stresses the importance of magnetic-dipole and g -factor measurements: not only can nucleon configurations be deduced, but any deviations from expected values from within an isotope chain can point towards structural transformations of the nucleus.

For specific odd-odd nuclei whose valence proton and neutron couple to form a state with $I \neq 0$, the empirical g factor can be calculated using the additivity relation:

$$g = \frac{1}{2} \left[g_p + g_n + (g_p - g_n) \frac{j_p(j_p + 1) - j_n(j_n + 1)}{I(I + 1)} \right], \quad (2.17)$$

as outlined in Ref. [Ney03], where g_p , g_n , j_p , and j_n represent the single-particle g factors and spins. Such values can be taken from either the (effective) Schmidt moments defined in (2.16), or from odd-even/even-odd nuclei with established single-particle configurations in the vicinity of the isotope in question. Deviations of experimentally obtained g factors from those calculated empirically with (2.17) can suggest configuration mixing. For two nuclear states $|j_1, I\rangle$ and $|j_2, I\rangle$ which contribute to the wavefunction Ψ , the magnetic-dipole moment μ_Ψ is given by:

$$\begin{aligned}
|\Psi\rangle &= a |j_1, I\rangle + b |j_2, I\rangle, \\
\mu_\Psi &= \langle \Psi | \boldsymbol{\mu} | \Psi \rangle = a^2 \langle j_1, I | \boldsymbol{\mu} | j_1, I \rangle + b^2 \langle j_2, I | \boldsymbol{\mu} | j_2, I \rangle + 2ab \langle j_1, I | \boldsymbol{\mu} | j_2, I \rangle,
\end{aligned}
\tag{2.18}$$

where a and b represent the respective contributions of the states defined above to μ_Ψ , on the condition that they are real values and they satisfy an $a^2 + b^2 = 1$ normalisation condition. While the first two matrix elements associated with the a^2 and b^2 terms simply represent the individual magnetic-dipole moments for the j_1 and j_2 states respectively, the final term in (2.18) represents a transition moment, which is non-zero only if the two states involved can be connected by an $M1$ transition. Spin-orbit partners with the same l but with $j_{1,2} = l \pm 1/2$ represent such an instance; associated ab contributions of only a few % can be sufficient to largely influence the total magnetic-dipole moment [Ari54; Bli54]. An in-depth configuration-mixing analysis involving these type of admixture contributions for the odd-odd isotope ^{214}Fr can be found in Appendix A.4.

The electric-quadrupole moment

Additionally, the electric-quadrupole moment of a nucleus is used to define its charge distribution symmetry. Under Cartesian coordinates, the operator Q is defined as:

$$Q = \sum_i^A q_i (3z_i^2 - r_i^2), \tag{2.19}$$

or alternatively using spherical tensors:

$$Q = \sqrt{\frac{16\pi}{5}} \sum_i^A q_i r_i^2 Y_2^0(\theta_i, \phi_i), \tag{2.20}$$

where q_i represent the effective proton and neutron charges used to take into consideration core-polarisation effects. Typical values are $q_\pi \sim 1.1 - 1.5$ and $q_\nu \sim 0.1 - 1.0$; see Table 2.1 for those associated with the LSSM calculations used in this thesis. From the $(3z_i^2 - r_i^2)$ term in (2.19), it is easy to see how the shape of the nucleus influences the electric-quadrupole moment value. Isotopes with a

zero electric-quadrupole moment are spherically symmetric, which is expected for closed-shell magic nuclei under the shell model framework. Positive and negative values correspond to prolate and oblate shapes respectively, along the direction of the z -axis. Likewise with the magnetic-dipole moment procedure in (2.14), experimental spectroscopic quadrupole moments Q_s are obtained from the expectation value of (2.20) under an $M = I$ projection:

$$\begin{aligned} Q_s &= \langle I, M = I | \mathbf{Q}_z | I, M = I \rangle \\ &= \sqrt{\frac{I(2I-1)}{(I+1)(2I+1)(2I+3)}} \langle I || \mathbf{Q} || I \rangle, \end{aligned} \quad (2.21)$$

which are zero for any nuclear states with $I \leq 1/2$, regardless of any deformation it may possess. As a consequence, the core has a zero electric-quadrupole moment.

Electric-quadrupole moments for isotopes near magic shell closures should in principle be determined by their valence-particle configurations. Associated single-particle values Q_{sp} for example, can be related to a particle/hole residing in a j orbital [Cas90]:

$$Q_{sp} = -q_j \frac{2j-1}{2j+2} \langle r_j^2 \rangle, \quad (2.22)$$

where $\langle r_j^2 \rangle$ represents the associated mean-square charge radius for that orbital. With respect to the nuclear shape as a whole, the negative-value prediction for a single valence particle from (2.22) can be interpreted as the polarisation of an otherwise spherical nucleus towards an oblate shape; this is *vice versa* for a single valence hole which corresponds with prolate deformation. Numerically evaluating (2.22) however, is a difficult task because the aforementioned variable requires a thorough understanding of the wavefunction (and the choice of mean field used) for the particle/hole. Besides, this extreme single-particle-model version is mostly never applicable to observed electric-quadrupole moments across the nuclide chart.

For an orbital containing more than one nucleon, the n particles under a seniority⁷ $\nu = 1$ scheme (with one unpaired nucleon) all contribute to Q_s according to:

⁷Seniority ν represents an additional quantum number to j , l , and s , which represents the degree of unpaired particles residing in a (part-)filled orbital with quantum numbers j , l ,

$$Q(j^n) = Q_{sp} \frac{2j+1-2n}{2j-1}, \quad (2.23)$$

as documented in Ref. [Ney03]. Looking at an isotope chain with even- Z where a neutron orbit is gradually filled for example, (2.23) should be expected to vary linearly between Q_{sp} ($n = 1$) and $-Q_{sp}$ (one hole in the orbital j) by varying odd- N , with a half-filled orbital expected to result in a zero electric-quadrupole moment. Such linearity can be observed for the odd- A ${}_{20}\text{Ca}$ ground-state isotopes residing in the $\nu 1f_{7/2}$ and $\nu 2p_{3/2}$ [Gar15b], as well as the odd- A ${}_{48}\text{Cd}$ isotopes (either the ground or isomer states) within the $\nu 1h_{11/2}$ orbital [Yor16]; any deviations point towards either core-contributions or signs of nuclear deformation. Interestingly, the removal of neutron pairs from an isotope chain with odd- Z and even- N induces a polarisation to the core which subsequently changes the electric-quadrupole moment, even though neutrons are not able to directly contribute. This effect is quadratic, resulting in parabolic trends between two magic shell closures which are maximal at the mid-shell; this is observed for the odd- A ${}_{29}\text{Cu}$ isotope chain between $N = 28 - 40$ and $N = 40 - 50$ [Gro17a]. Considering that the electric-quadrupole moments for j_π one-particle and one-hole systems are related to each other according to:

$$Q_{sp}(j_\pi^1) = -Q_{sp}(j_\pi^{-1}), \quad (2.24)$$

it follows that the parabolic trends formed from odd- Z isotope chains which possess either one proton-particle or one proton-hole in an orbital j , are expected to be mirror opposites of one another. One example involves ${}_{29}\text{Cu}$ and ${}_{31}\text{Ga}$, in which their respective one-proton and one-hole systems originate from the $\pi 2p_{3/2}$ orbital; see Chapter 5.4 for further details.

The mean-square charge radius

Characterisation of the nuclear radius is fundamentally important for defining the size of a nucleus. A variety of experimental methods are available, however the type of radii extracted depends on the choice of probe: while a hadronic probe would result in the *matter radius*, the use of an electromagnetic-type probe would only be sensitive to the charged protons, yielding the *charge radius*

and 1 [Rac43; Ney03]. It is indicative of deviations from single-particle behaviour, because states with $\nu > 1$ typically have $I \neq j$. Further elaboration is given in Chapter 6.1 in which it is needed to explain the ground state of ${}^{79}\text{Ga}$.

instead. The difference between the two radii is ordinarily small for isotopes residing on or close to the stability line; this generally increases with $(N-Z)/A$. Exceptionally large differences can be prevalent in very neutron-rich species, such as halo nuclei [Han87; AIK96].

Early descriptions of the nuclear size assumed spherical Liquid-Drop (LD) shapes with a radius R , proportional to its mass A :

$$R = R_0 A^{1/3}, \quad (2.25)$$

where $R_0 \approx 1.2 - 1.25$ fm. As the proton and neutron distributions have a similar spatial extension in most nuclei, the proton-charge distribution is considered a good probe for the size of a nucleus. Quantum mechanically, the nuclear-charge/proton-charge distribution $\rho_{n/ch}$ of the nucleus is defined as [Fri04]:

$$\begin{aligned} \langle r^N \rangle &= \frac{\int \rho_n(\mathbf{r}) r^N d\mathbf{r}}{\int \rho_n(\mathbf{r}) d\mathbf{r}}, \\ \langle r_{ch}^2 \rangle &= \frac{\int \rho_{ch}(\mathbf{r}) r^2 d\mathbf{r}}{\int \rho_n(\mathbf{r}) d\mathbf{r}}, \end{aligned} \quad (2.26)$$

where the denominator in both expressions is equivalent to the total nuclear charge: eZ . The choice of looking at $\langle r_{ch}^2 \rangle$ in (2.26) is deliberate because it is the only term which laser spectroscopy methods can directly probe. Furthermore, only the differences between two isotopes A and A' can be extracted, which are normally denoted as $\delta\langle r^2 \rangle^{A,A'}$; the ‘ ch ’ subscript is now disregarded for the rest of this thesis.

Any deviations of the mean-square charge radius from its spherical prediction $\langle r_{sph}^2 \rangle$ can indicate a deformed nucleus. With respect to $\langle r^2 \rangle$ and $\delta\langle r^2 \rangle^{A,A'}$, these relations for axially-symmetric nuclei are given by [Nör14]:

$$\begin{aligned} \langle r^2 \rangle &= \langle r_{sph}^2 \rangle \left(1 + \frac{5}{4\pi} \langle \beta_2^2 \rangle \right) + 3\sigma^2, \\ \delta\langle r^2 \rangle^{A,A'} &= \delta\langle r_{sph}^2 \rangle^{A,A'} \left(1 + \frac{5}{4\pi} \delta\langle \beta_2^2 \rangle^{A,A'} \right), \end{aligned} \quad (2.27)$$

where σ represents the nuclear surface diffuseness parameter which, irrespective of the nuclear system, is constant. This explains its absence from the latter expression in (2.27). From the mean-square quadrupole deformation parameter $\langle\beta_2^2\rangle$, it is easy to see how an increased degree of nuclear deformations can correspond to a larger mean-square charge radius for a given isotope. Values for $\langle r_{sph}^2\rangle$ in (2.27) can originate from LD model estimations [Boh98]:

$$\langle r_{LD}^2\rangle = \frac{3}{5}R^2 = \frac{3}{5}R_0^2A^{2/3}, \quad (2.28)$$

using the same parameters as defined for (2.25). Alternatively, other theoretical droplet-model interpretations are available which are still often used to interpret extracted mean-square charge radii [Mye74; Mye83; Ber85].

Interesting features can also be exhibited when examining $\langle r^2\rangle$ values across an entire isotope chain. A sudden, distinct discontinuity in its slope is usually indicative of crossing a magic shell closure. More commonly referred to as a ‘kink’, such occurrences have been observed within many isotope chains for several well-established magic numbers across the nuclide chart [Ang13; Cam16]. This, within the context of the shell model, can be explained by the larger radial extent of the next available shell-model orbital, in which the valence neutron resides. As recently explained in Ref. [Gor19], the attractive nature of pn interactions causes an expansion of the proton distribution within the nucleus which reduces the overall symmetry energy. All of this can be related to the spin-orbit interaction [Rei95; God13; Nak15], which has already explained the kink at $N = 126$ in the $_{82}\text{Pb}$ isotope chain with its inclusion in Relativistic Mean-Field (RMF) models [Sha95; God13]. Other nuclear effects arising from: pairing [Taj93; Fay96], ground-state correlations [Ben06], and compressibility [Nie12], are suggested to also contribute towards the increased magnitude of these kinks at magic shell closures.

Additionally for (even- Z and odd- Z) isotope chains, the charge radius for a particular even- N species is in general larger than the average of its odd- N neighbour species. This phenomena is known as the Odd-Even Staggering (OES) effect and it arises from a suppression of pair scattering by the unpaired neutron in the odd- N species [Zaw85; Rei17]. Thus, OES effects are indicative of any proton-excitation and pn -correlation variations along a particular isotope chain. Cases for the $_{20}\text{Ca}$, [Gar16; Mil19] $_{29}\text{Cu}$ [Bis16], $_{30}\text{Zn}$ [Xie19], $_{80}\text{Hg}$ [Mar18a] isotope chains can be found in the corresponding references. An overview for the isotope chains residing in the near vicinity of $Z = 82$ and the doubly-magic ^{208}Pb core, can be found in Ref. [Coc17].

Isomeric states

Experiments to determine such properties defined above are not solely limited to the ground state of a nucleus: studies on isomeric states are just as relevant. Instances with lifetimes that far exceed the usual ps timeframe, usually at least a factor 10^3 longer, are considered isomer states and are commonly denoted with an m after the exponent mass number: ^{239m}Pa for example, the first known isomer discovered in 1921 [Hah21]. The range of observed durations is additionally rather dramatic: from ns (^{196m}Pb ; $t_{1/2} = 9.4(5)$ ns) [Bal02], to hours (^{85m}Kr ; $t_{1/2} = 4.480(8)$ h [Woh70]) to $\geq$ Gyr (^{180m}Ta ; $t_{1/2} > 10^{16}$ years [Hul06]). In particular for the latter example, the requirement of an $M8$ ($9^- \rightarrow 1^+$) transition to the ground state explains its practically-stable nature. With respect to laser-spectroscopy techniques, only long-lived isomer states typically with lifetimes at least of the $\sim$ ms order can be studied; see Chapter 7.1.2 for measurements of ^{214g}Fr with $t_{1/2} = 5.0(2)$ ms, and Ref. [Moo02] for such studies on ^{130m}Ba with $t_{1/2} = 9.54(14)$ ms. Although the major limitations are borne from production and measurement techniques, shorter-lived nuclear states down to $\sim \mu\text{s}$ have been probed with laser-spectroscopy techniques in dedicated studies, such as ^{85m}Rb with $t_{1/2} = 1 \mu\text{s}$ [Shi84].

Isomeric states are often peculiarities within the realms of nuclear structure and provide motivation for nuclear-structure studies to fully investigate how these manifest. While shell-model principles and Paar's parabola rule (see Chapter 7.2.1) can be utilised to explain their occurrences, obtaining a complete insights may not always be easy or obvious. Long-lived high-spin isomeric states can also result if an excitation of the nucleus induces the break-up of a nucleon pair, which then couples to the highest J -values as allowed from (2.7). Special mention is given to the trans-lead region in which high- j single-particle orbitals can potentially be occupied. Such examples include ^{203}Pb [Lin77], ^{212}Po [Pol87], and ^{215}Bi [Kur03].

2.2 Elements of atomic physics

The inclusion of every interaction between the nucleus and the orbiting electrons is essential for an accurate description of the atomic system. Fine structure for example, requires accounting for: the Coulomb interaction, the relativistic Schrödinger equation, the spin-orbit interaction, the Darwin term, and the Lamb shift. Additional electromagnetic and finite size corrections of the nucleus inside the atom lead to the manifestation of hyperfine structure which if measured accurately for a nuclear system, yields the extraction of important structural properties which are essential for independently testing nuclear models.

2.2.1 Hyperfine structure

The Coulomb interaction is responsible for the fine structure of a particular element with Z electrons, whose electronic quantum levels are usually characterised by the electronic spin $\mathbf{J}$. Shifts in these levels occur due to the interaction of the electron cloud with the nuclear charge distribution and total nuclear spin $\mathbf{I}$, ultimately revealing a non-degenerate hyperfine structure with levels defined by a total angular momentum $\mathbf{F}$:

$$\mathbf{F} = \mathbf{I} + \mathbf{J}, \quad (2.29)$$

with the application of (2.7). Overall, the repertoire of allowed hyperfine levels goes from $I + J$ to $|I - J|$, in integer steps. The manifestation of hyperfine structure originates from the interactions between the total field generated by the orbital electrons and the multipole moments of the nucleus. With respect to the total Hamiltonian, its potential H can be described by:

$$H = H_{E0} + H_{M1} + H_{E2} + H_{M3} + \dots, \quad (2.30)$$

where the subscripts denote all allowed multipole contributions.

Electric-monopole ($E0$) contributions

The electronic level ordering of an atomic system is borne from the Coulomb interaction between the electron cloud and a point-charge nucleus with charge eZ , which can be considered as an electric-monopole interaction [Dra06]. This is purely Coulomb in nature and can be related to:

$$H_{E0} \propto V_{Coulomb} = \frac{Ze^2}{4\pi\epsilon_0 r}, \quad (2.31)$$

where Z is the proton number (and thus the number of electrons in a neutral atom), e is the electron charge, ϵ_0 is the permittivity of free space, and r is the distance between the electron and the nucleus. Interestingly, shifts in these levels are seen across isotopic chains as a function of N . This is because the nucleus is not a point-charge and its charge distribution changes with the number of neutrons in a nucleus. Between any two given isotopes, this is more commonly known as the *isotope shift* and is explained in more detail in Chapter 2.2.2.

Magnetic-dipole ($M1$) contributions

Interactions between the magnetic-dipole moment of the nucleus, $\boldsymbol{\mu}$, with the magnetic field generated by the electrons, $\mathbf{B}_0$, lead to the occurrence of the hyperfine levels given in (2.29) for all nuclei with $I, J > 0$. The magnitude of this hyperfine-level shift with respect to the fine-structure level J , for a given F state is:

$$\begin{aligned} H_{M1} &= -\boldsymbol{\mu} \cdot \mathbf{B}_0 = -\frac{\mu B_0}{IJ} \mathbf{I} \cdot \mathbf{J} \\ &= -\frac{1}{2} \frac{\mu B_0}{IJ} (F(F+1) - I(I+1) - J(J+1)) \quad (2.32) \\ &= -\frac{1}{2} AK, \end{aligned}$$

where the expression $\mu B_0/IJ$ is commonly referred to as the A -hyperfine parameter. The latter form in (2.32) is most commonly used and simply describes this Hamiltonian contribution as a product of the nuclear magnetic-dipole moment and atomic magnetic field (μB_0), with a numerical factor (K) describing the orientation of the nuclear spin I , relative to the electronic spin J .

Electric-quadrupole ($E2$) contributions

For $I, J > 1/2$, hyperfine levels are further shifted by interactions between the spectroscopic electric-quadrupole moment of the nucleus, Q , and the electric field gradient generated by the electrons, V_{zz} . This contribution can be quantified by:

$$\begin{aligned} H_{E2} &= eQV_{zz} \cdot \frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)} \\ &= B \frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)}, \end{aligned} \quad (2.33)$$

where K has already been defined in (2.32) and the expression eQV_{zz} is denoted as the B -hyperfine parameter. Analogous to (2.32), (2.33) can be expressed as a product of physics observables with the angular-momentum orientation contribution.

Higher order contributions

Other multipole contributions listed in (2.30), such as the magnetic-octupole ($M3$) interaction and other higher-order varieties, can induce additional shifts to the hyperfine structure. However, the size of these effects decreases substantially with increasing order; for most laser-spectroscopy studies, only ($M1$) and ($E2$) contributions are considered. Nonetheless, magnetic-octupole moments have been studied for isotopes, albeit only very few [Dal54; Ger03; Sin13].

2.2.2 Isotope and isomeric shift

As introduced in Chapter 2.2.1, differences in the electronic-level energies occur between species in an isotope chain with the neutron number N . Consequently, their centroid frequencies ν also vary whereby the difference between two isotopes with masses A and A' :

$$\delta\nu^{A,A'} = \nu^{A'} - \nu^A, \quad (2.34)$$

is known as the *isotope shift*. More explicitly, this quantity is made up of two additive contributions which are directly related to the neutron number N :

- Firstly, the difference in N induces changes in the nuclear charge distribution and thus, to the charge radius. As a result, the electrons feel different electrostatic potentials, leading to a frequency shift referred to as the *field shift*⁸:

$$\delta\nu_{FS}^{A,A'} = \frac{Ze^2}{6h\epsilon_0} \delta|\Psi(0)|^2 \delta\langle r^2 \rangle^{A,A'} = F \delta\langle r^2 \rangle^{A,A'}, \quad (2.35)$$

which is directly related to the change in the mean square charge-radius, $\delta\langle r^2 \rangle^{A,A'}$. The $\delta|\Psi(0)|^2$ term represents the change in the electron-cloud density at the site of the nucleus between the initial and final states of the electron transition; along with the $Ze^2/6h\epsilon_0$ term, it is usual to encapsulate this combined expression with the field-shift factor F , shown at the end of (2.35).

- Secondly, the mass difference between the isotopes, characterised by the reduced mass of the combined system, has an additional effect on the fine structure levels and is known as the *mass shift*:

⁸It is this contribution to the isotope shift that is directly relatable to the electric monopole ($E0$) interaction.

$$\delta\nu_{MS}^{A,A'} = M \frac{m_{A'} - m_A}{m_A m_{A'}}, \quad (2.36)$$

where m represents the masses of isotopes with A and A' , and M is known as the mass shift factor. With an approximate m^{-2} dependence, this contribution to the total isotope shift becomes more negligible for the heavier isotope species; approximate equivalence between the mass and field shift contributions occurs at $Z \sim 38$ [Nör14].

Overall for (2.34):

$$\delta\nu^{A,A'} = \delta\nu_{FS}^{A,A'} + \delta\nu_{MS}^{A,A'} = F\delta\langle r^2 \rangle + M \frac{A' - A}{AA'}, \quad (2.37)$$

in which the atomic (F , M) and nuclear (all other terms) dependencies for the isotope shift can be clearly identified. For ground and isomeric states within the same nuclear system, (2.37) can instead yield the *isomeric shift*. With zero mass shift present, they present ideal probes for mass-shift-independent determinations of F for a particular transition; see Ref. [Yor16] for an example application for the $_{48}\text{Cd}$ isotope chain.

2.3 The concepts of laser spectroscopy

Optical spectroscopy is by no means a novel technique. As early as the 1960's, such methods regularly probed the hyperfine interaction described in Chapter 2.2.1 for many stable and long-lived radioactive species with light from a flashlamp or an interferometer [Jac79]. Nowadays, laser light is used to irradiate an isotope of interest with a spin I ; its frequency ν_0 depends on the fine structure transition under investigation characterised by $J \rightarrow J'$ from the spectroscopic⁹ terms. By scanning over a certain frequency range with respect to the nuclei rest frame, hyperfine transitions are probed whenever the frequency matches an electron transition between a lower (l) F level and an upper (u) F' level with a frequency difference analogous to (2.30):

⁹Levels forming the isoelectric sequence of a particular element are usually written in spectroscopic notation as $n^{2S+1}L_J$. Usually, it concisely describes the overall configuration of any valence electrons present using their J , L , and S quantum numbers/notations, while the configuration of the other innermost electrons are usually expressed in a contracted n form. For example, the full ground-state notation of atomic gallium is $[\text{Ar}]3d^{10}4s^24p^2P_{1/2}$.

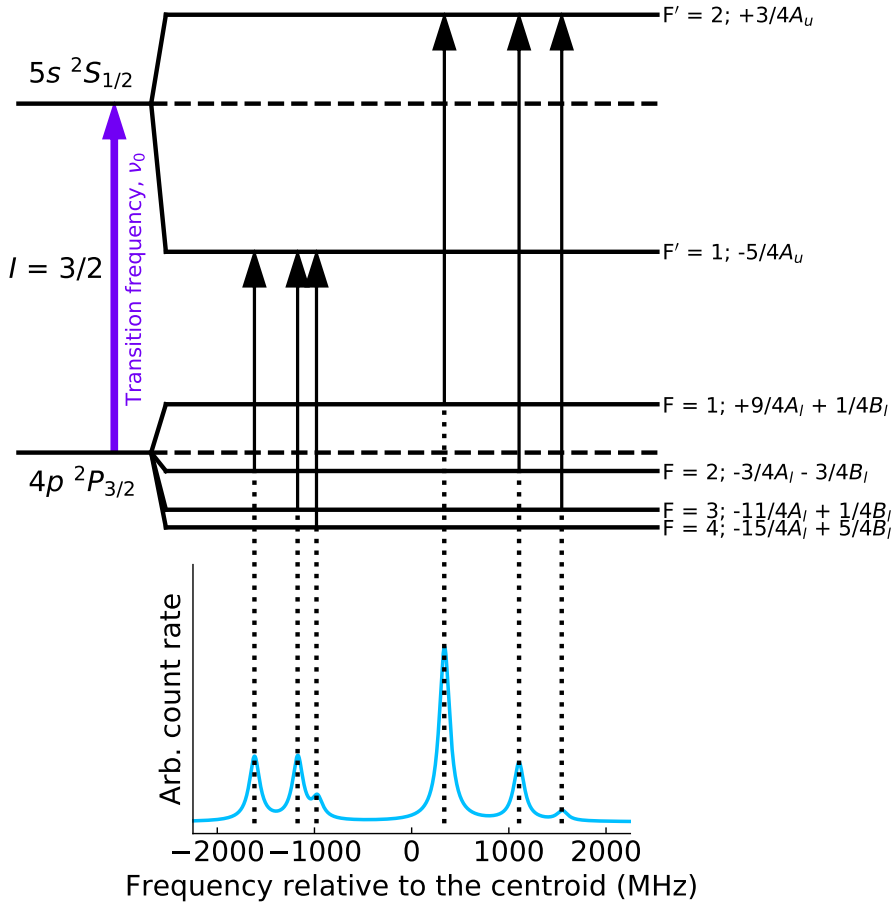


Figure 2.3: Allowed hyperfine transitions for the $4p \ ^2P_{3/2} \rightarrow 5s \ ^2S_{1/2}$ transition in ^{71}Ga with $I = 3/2$, along with their appearance in a simulated experimental hyperfine spectrum. Each hyperfine level is labelled with its perturbation from the corresponding lower (l) or upper (u) fine structure level.

$$\begin{aligned} \Delta\nu^{F,F'} = \nu_0 + \frac{K'}{2}A_u + \frac{3K'(K'+1) - 4I(I+1)J'(J'+1)}{8I(2I-1)J'(2J'-1)}B_u \\ - \frac{K}{2}A_l - \frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)}B_l, \end{aligned} \quad (2.38)$$

incorporating up to $E2$ -type contributions as discussed in Chapter 2.2.1. Selection rules determine which hyperfine transitions can be allowed, which depends on the quantity of angular momentum exchanged between the two fine-structure levels: $\Delta J = |J - J'|$. For the majority of laser spectroscopy studies (including the transition featuring in Figure 2.3), $\Delta J = 1$ and thus, the electric-dipole selection rules are enforced:

$$\Delta F = 0, \pm 1, \quad \text{and} \quad F = 0 \not\rightarrow F' = 0. \quad (2.39)$$

It is typical to observe several hyperfine resonances in a given spectrum, with its quantity generally increasing for higher values of I and/or J and J' . For example in Figure 2.3, six resonances are observed for ^{71}Ga using the $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transition. By identifying them all, the extraction of the A - and B -hyperfine parameters for both states (where applicable) and the isotope shift is possible, from which fundamental ground-state properties such as: μ , Q_s , I , and $\delta\langle r^2 \rangle$, can be extracted. The elegance of these physics concepts requires no nuclear-model dependencies whatsoever and thus, comparisons with the aforementioned ground-state properties theoretically predicted with, *e.g.* shell-model interactions presented in Chapter 2.1.1, provide useful tests of validity.

2.3.1 Extracting the electromagnetic moments

With the connection between A and the magnetic-dipole moment μ already established in (2.32), the only variable that needs evaluating is B_0 . It is customary to assume this parameter is constant across an isotope chain; by taking the ratio of two A -hyperfine parameters from different isotopes:

$$\mu_m = \mu_{ref} \frac{I_m A_m}{I_{ref} A_{ref}}, \quad (2.40)$$

where the subscripts *ref* and *m* refer to a reference isotope and an isotope under measurement, respectively. Caution must be exercised regarding any non-uniformities present in the extended nuclear magnetisation and charge distribution resulting from nuclear-structure changes between isotopes. Known as the Bohr-Weisskopf and Breit-Rosenthal-Crawford-Schawlow effects respectively [Boh50; Ros32; Cra49], together they correct for the point-like-nuclei assumption in (2.40) according to:

$$\mu_m = \mu_{ref} \frac{I_m A_m}{I_{ref} A_{ref}} (1 + {}^{ref}\Delta^m), \quad (2.41)$$

where ${}^{ref}\Delta^m$ is known as the hyperfine anomaly between two given isotopes. Sensitivity to the hyperfine anomaly usually requires high-precision experiments for the A -hyperfine parameters, with Ref. [Sch18] providing an example needed for the ${}^{208,209}\text{Bi}$ isotope pair. A current up-to-date compilation of observed hyperfine anomalies in nuclear systems can be found in Ref. [Per13]. While for gallium it can be considered as a negligible $\sim 10^{-3}$ effect, it is more prevalent in francium isotopes of up to a $\sim 1\%$ contribution (see also Refs. [Zha15; Wil17a]). Dealing with instances of the latter normally involves including these effects as an additional uncertainty contribution to the total magnetic-dipole moment.

With respect to the B -hyperfine parameter and the electric-quadrupole moment in (2.33), only V_{zz} needs to be defined. As the electronic cloud is the same for all isotopes, it is invariant throughout an isotopic chain and by taking the ratio of two B -hyperfine parameters from different isotopes:

$$Q_{s,m} = Q_{s,ref} \frac{B_m}{B_{ref}}, \quad (2.42)$$

using the same subscript definitions as in (2.40). Reference spins, hyperfine parameters, and electromagnetic moments required for (2.40) and (2.42) should ideally come from isotopes which have been measured precisely. Depending on the available measurements for a given isotope chain, chosen references can come from either a predetermined isotope from the laser spectroscopy experiment, or other independent measurements such as those from atomic-beam or β -Nuclear Magnetic Resonance (β -NMR) spectroscopy.

As alluded in Chapter 2.1.2, extraction of the magnetic-dipole and electric-quadrupole moments requires at least one level in the transition scheme possessing $J \geq 1/2$ and $J \geq 1$ respectively. Although a greater sensitivity can generally be achieved with larger state splittings, this is dependent on the relative strength between the associated A - and B -hyperfine parameters for the two states involved.

2.3.2 Extracting the changes in the mean-square charge radii

In order to extract changes in the mean-square charge radii from the measured isotope shifts using (2.37), knowledge of the atomic parameters F and M

are needed for a particular transition. Although semi-empirical [Blu87] and MultiConfiguration Dirac-Fock (MCDF) [Fri12] calculations are able to provide theoretical estimates, many difficulties can arise due to their complexities [Che12b]. However if F and M are known for some other transition, then a King plot method can be utilised to determine these parameters for the probed transition [Kin84]. Considering $\delta\langle r^2 \rangle$ remains invariant for all possible transitions of an element, (2.37) can be equated for any two transitions labelled 1 and 2:

$$\delta\langle r^2 \rangle^{A,A'} = \frac{1}{F_1} \left(\delta\nu_1^{A,A'} - M_1 \frac{m_{A'} - m_A}{AA'} \right) = \frac{1}{F_2} \left(\delta\nu_2^{A,A'} - M_2 \frac{m_{A'} - m_A}{AA'} \right), \quad (2.43)$$

which provides the following relation between the two isotope shifts:

$$\mu_{A,A'} \delta\nu_1^{A,A'} = \frac{F_1}{F_2} \mu_{A,A'} \delta\nu_2^{A,A'} - M_2 \frac{F_1}{F_2} + M_1, \quad (2.44)$$

where $\mu_{A,A'} = \frac{m_{A'} m_A}{(m_{A'} - m_A)}$ denotes a mass modification factor for the two isotopes concerned. The consequence of (2.44) means that plotting $\mu_{A,A'} \delta\nu_2^{A,A'}$ versus $\mu_{A,A'} \delta\nu_1^{A,A'}$ should display a simple linear relationship, with a gradient and intercept of F_1/F_2 and $-\frac{F_1}{F_2} M_2 + M_1$ respectively. Atomic information for both transitions must not only be well known, but also comprise from a minimum of three isotopes [Coc17]. While this is not a problem for most even- Z elements with $Z \leq 82$, which possess several stable isotopes, further difficulties arise with odd- Z elements which mostly have only one or two stable isotopes, or wholly radioactive species. It may be necessary for these latter cases to include non-optical data in the King plot analysis, such as from either: theoretical calculations, electron-scattering experiments, or experimental muonic X-ray data [Fri04].

Choosing transition schemes which are sensitive to charge-distribution changes are of utmost importance for the successful extraction of isotope shift data. Non-zero F -atomic factors from (2.35) are obtained when there is a significant change in the electron-wavefunction overlap with the nucleus (*id est* from the $\delta|\Psi(0)|^2$ term), implying that electronic s and/or $p_{1/2}$ occupancies should preferentially be chosen. In most cases, $s \rightarrow p$ transitions are studied.

2.3.3 Extracting the nuclear spin

A combination of approaches are available for measuring the spin assignments of isotopes, with Figure 2.4 serving as further graphical enlightenment for the discussions presented in this section.

The simplest method is to merely count the number of observed resonances. From (2.29), any given J level splits into $2J + 1$ hyperfine substates for $I \geq J$; progressively fewer splittings arise with lower I values, with $I = 0$ resulting in a single unperturbed level. As a consequence, nuclear structures with $I = 0 - J_m$, where J_m is equal to the greater J value involved, possess a unique number of (up to $2I + 1$) allowed hyperfine transitions and such an example can be seen in Figure 2.4(a). Arguably this is the most unequivocal method for the assignment of nuclear spins; further sensitivity can be achieved by choosing transition schemes with the highest possible J values. However this can also prove counterintuitive, since an increased number of allowed transitions with

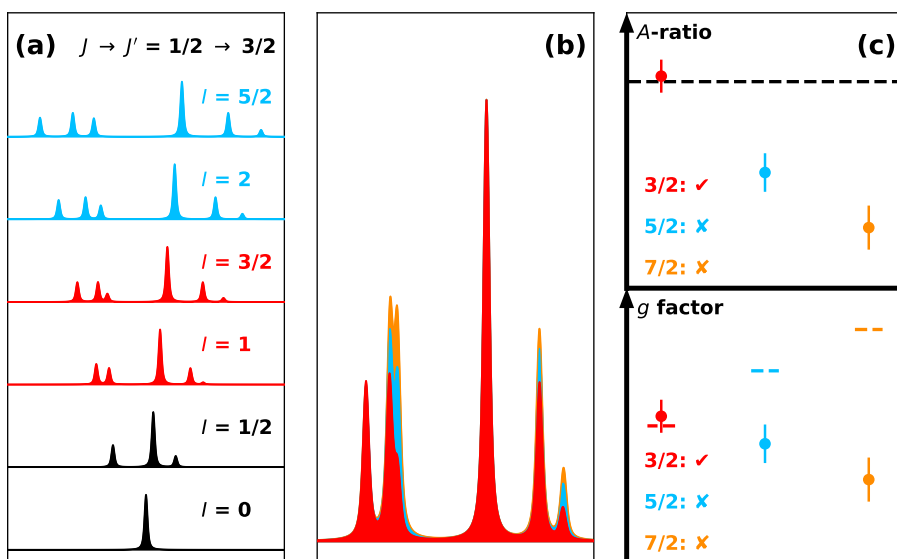


Figure 2.4: Available methodologies for determining the nuclear spin in laser spectroscopy. Using a $J = 1/2 \rightarrow J' = 3/2$ transition scheme as an example: (a) all structure with spin assignments up to and including $I = 3/2$ comprise of a unique number of resonances. For higher spin values, comparisons between: (b) the peak intensities, and (c) the extracted A -hyperfine ratios and/or calculated g factors, can be utilised instead to infer spin assignments.

a greater overlap, can potentially make searches rather cumbersome. Coupled with a generally greater intensity variation between the peaks, this can make it incredibly challenging to distinguish the least intense transitions from any present background level. It is stressed that the right compromise should be used according to how effectively these complications can be managed. This framework is further discussed with respect to a variety of atomic gallium transition schemes in Chapter 8.1.

Alternatively, the peak heights of every hyperfine component in a given spectrum $S_{FF'}$, can be compared. Theoretically, they can be related to the intensity of the underlying fine structure transition $S_{JJ'}$ [Bla13], by:

$$\frac{S_{FF'}}{S_{JJ'}} = (2F + 1)(2F' + 1) \left\{ \begin{matrix} F & F' & 1 \\ J' & J & I \end{matrix} \right\}, \quad (2.45)$$

where the expression enclosed within the brackets is the Wigner-6j symbol. Calculated relative intensity differences for each $F \rightarrow F'$ transition assuming different I values can be compared to those experimentally observed; Figure 2.4(b) highlights how the peak intensities may differ for a variety of I . However, care should be taken regarding saturation and optical pumping effects which can alter the peak intensities with respect to each other [Gro17d], as well as an increasing indistinguishability for higher values of I .

For scenarios in which the hyperfine anomaly in (2.41) can be specifically neglected, the ratio of A -hyperfine parameters between the two atomic states in an isotope should remain constant for all isotopes. Only the correct I value results in the correct A -ratio, with Figure 2.4(c) showing how this can be inferred amongst other spin assignments which produce different ratios. For nuclear states that are known to be of a dominantly single-particle nature, a comparison of the resulting g factors with those empirically calculated from (2.16) or (2.17) can also provide further confirmation in addition to the structural insights it already brings. However these methodologies can prove inconclusive when higher values of spin are involved [Lyn16], highlighting the need for additional confirmatory methods such as a decay-spectroscopy analysis.

It is remarked that while the labelling of nuclear states (where applicable) combine the spin and parity together, as I^π , laser-spectroscopy techniques cannot be used to discern the parity directly. Thus, the extraction of spin values from hyperfine-spectroscopy measurements displayed in this thesis involve just the I value. It is possible for parity to be implied however, whenever shell-model insights are used to interpret extracted nuclear-physics observables. Comparisons of g factors with: effective single-particle estimates, pn configurations, or LSSM calculations, all represent appropriate examples, the latter in particular having

been utilised to solve a previously contentious debate of the $^{72,74}\text{Cu}$ ground states [Fla10]. If a clear distinction in this thesis can be made, nuclear-state parities are also suggested in any subsequent discussion.

2.3.4 Summary

A robust theoretical framework is usually necessary for explaining phenomena that result from experimental observations. Without it, one may as well be searching in the darkness for explanations. Thankfully the nuclear shell model provides a simple, yet powerful methodology for describing the structural characteristics of the nucleus. Important parameters include: the magnetic-dipole and electric-quadrupole moments, changes in the mean-square charge radii, and the nuclear spin, all of which are easily accessible using laser-spectroscopy techniques. Overall, these provide the necessary building blocks which are key for discussing the wider picture of changing nuclear-structure phenomena, exhibited across the nuclide chart.

Chapter 3

Experimental details for observing hyperfine structures

From the bygone age of relying predominantly on theoretical predictions, technological advances have made it easier than ever to directly probe physics using experimental techniques. Of course, one must choose an appropriate experimental setup when delving into the mysteries of nuclear physics. Measurements presented later in this thesis were obtained using the CRIS technique at the Isotope Separator On-Line DEvice (ISOLDE) facility in CERN. While a relatively new setup [Fla13a], it is currently at the forefront of cutting-edge laser-spectroscopy techniques, able to perform sensitive measurements on isotopes with exceptionally high efficiencies, amongst relatively low backgrounds. This next chapter will narrate the voyage for isotopes of interest from their inceptions at the ISOLDE facility, to their detection at the CRIS setup. Key concepts which help further extend the bounds of exoticity will also be discussed.

3.1 The ISOLDE facility at CERN

Situated on the outskirts of Geneva and home to currently the largest particle accelerator in the world, is CERN. Although recently synonymous with the famous ground-breaking Higgs Boson discovery within the realms of particle physics [Col12a; Col12b], one of its earliest facilities was the ISOLDE. Commencing in 1967 [Bor17], it is home to a smörgåsbord of experiments dedicated to nuclear-structure studies. Such examples include:

laser spectroscopy setups like COLLAPS [Gar16; Wra17] and CRIS [Lyn16; Gro17b] for measuring the ground-state properties of isotopes, the ISOL TRAP (ISOLTRAP) for nuclide mass measurements [Ata15; Mou18], the ISOLDE Decay Station (IDS) for investigating isotope decay properties [Kir18; Pie19], and the Weak Interaction Studies with ^{32}Ar Decay (WISArD) experiment for characterising weak interactions and constraining Standard-Model physics [Sev17].

Colloquially, ISOLDE is known as a radioactive *isotope factory* in the sense that a large swathe of over 1300 species from 71 elements of the periodic table can be accessed [Bor17] from the nuclide chart in Figure 1.1. Handily incorporated into

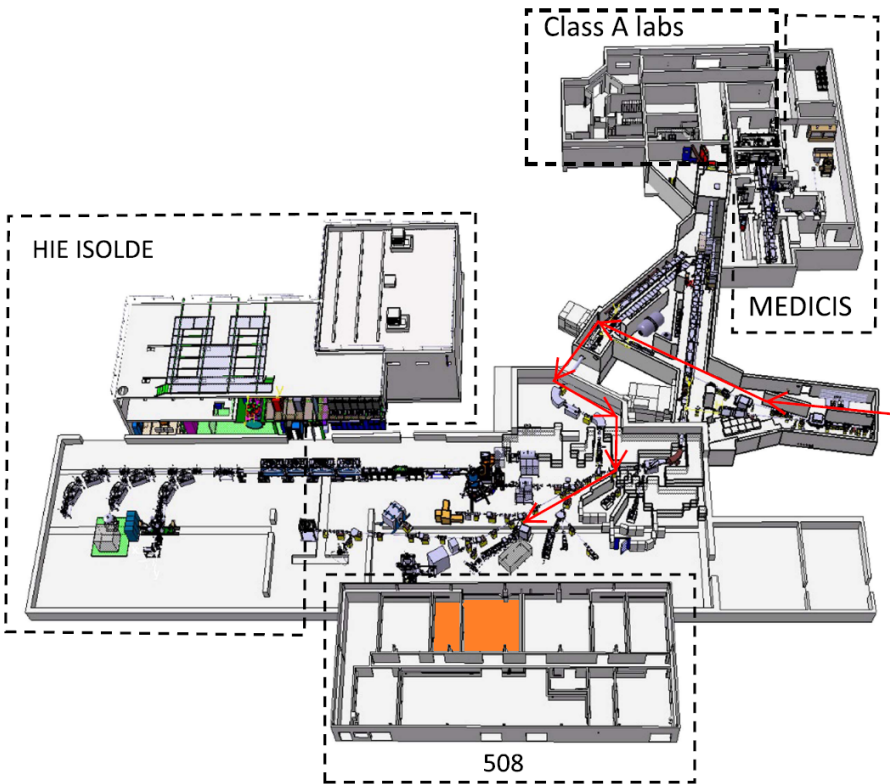


Figure 3.1: A schematic of the current layout of the ISOLDE facility. The red arrows highlight the journey of proton pulses and subsequent radioactive products through ISOLDE to the CRIS setup; see text for further details. The CRIS laser lab is highlighted in orange. Picture taken from Ref. [Cat17].

its acronym, ISOLDE utilises the Isotope Separation On-Line (ISOL) method in which a thick target is bombarded with a beam of protons. After diffusion and effusion of the (radioactive) isotopes out of the target during a $\sim$ ms timeframe, they are ionised and directly sent to an experimental setup. Figure 3.1 shows the complete layout of the ISOLDE complex with a red highlighted path which will be referred to in the following sequential description of its operations. The first red arrow depicts the entry of proton pulses into ISOLDE, originating from the Proton Synchrotron Booster (PSB): each accommodating of the order 10^{13} , with an energy and average beam intensity of 1.4 GeV and $2 \mu\text{A}$ respectively. They are delivered in 1.2 s intervals and usually organised into a regular sequence known as the proton supercycle, consisting of a mix of proton/blank pulses up to a total of 30 - 40 pulses, each separated by the aforementioned time interval. ISOLDE uses the majority of the protons handled by the PSB, taking a 61%¹ share in 2016.

3.1.1 Isotope production

Following up to the end of the second arrow in Figure 3.1, proton pulses impinge the target which produces an array of radioactive products from fission, spallation, and fragmentation reactions. Another target station is also available at ISOLDE which is situated towards the left at the end of the first arrow. Both offer particular advantages with the resulting isotope beam that can be delivered to an experimental setup and will be further discussed in Chapter 3.1.3. While uranium carbide (UC_x) is a popular target material choice for several experimental campaigns due to its versatility, over 25 different materials are available offering bespoke beam purities, production mechanisms and yields, and limits of exoticity [Her10]. UC_x targets are roughly 45 g/cm^2 thick; this quality is essential for maximising the number of nuclear interactions made within its material which occurs with approximately 10%² of the protons. Subsequent extraction and ionisation of the resulting products is dependent on the time required to effuse and/or diffuse out of the target, which is normally heated to temperatures $> 2000 \text{ K}$ to facilitate these processes. They are also highly correlated to the chemical properties of an element; the different release mechanics of the measurement species and any unwanted contamination can prove essential for suppressing the presence of the latter.

¹Numerically, this is equivalent to $\sim 8 \times 10^{19}$ protons. While this may seem sizeable, its combined weight is less than 1% of that of an ordinary human eyelash.

²Recently, the other 90% which traverse through the target have been utilised by the MEDical Isotopes Collected from ISolde (MEDICIS) facility. A second target has been placed after the target at the end of the second arrow in Figure 3.1 and is used to produce radioisotopes for medical research [San14; Cat17].

An external signal known as the *beam gate* can be utilised at ISOLDE to either prematurely block, or delay the release of the ion-beam signal into the ISOLDE beamline. Undertaking both actions may also be necessary. Example cases of operation will now be discussed for ^{214}Fr ($t_{1/2} = 5.0(2)$ ms [Tor68]) and ^{62}Ga ($t_{1/2} = 116.100(25)$ ms [Gri08]) in which the manipulation of the beam gate was necessary for optimising their measurements. Generally speaking for short-lived isotopes, it is usually prudent to restrict the temporal length of the beam gate (*id est*, how long to leave it open for) to a maximum of a few half-lives. After this time, any short-lived species produced upon the proton-beam impact would likely have wholly decayed and thus, continuing with any on-going measurement with an isotope beam purely consisting of contaminants would be counterproductive. For ^{214}Fr and ^{62}Ga , beam-gate lengths of ~ 35 - 50 ms and ~ 300 - 400 ms, from the moment of proton impact, have been sufficient to majorly limit the presence of isobaric radium and copper respectively, in the isotope beam. Delayed beam gates with respect to the proton-pulse impacts have also been implemented, however this is necessitated for different reasons concerning the cases of these two isotopes. For ^{214}Fr , it is needed because of its lifetime similarity with the high-voltage recovery time (~ 2 - 5 ms), whenever it switches off due to impending proton-pulse impacts. Neglecting this consideration can potentially interfere with the beam delivery and data acquisition used for measurements of short-lived species [Cat17]; thus, a ~ 5 - 10 ms delay is usually sufficient. While this does not present a problem for ^{62}Ga with its much-longer half-life, a delay is nonetheless needed because of the fast-release characteristic for isobaric zinc [Köe05]. From the aforementioned reference, delaying the beam gate by ~ 200 ms after proton-pulse impacts can suppress this contamination by up to a factor of 2 - 4. Choosing a particular temporal-beam-gate period may also be advantageous if an optimal isotope-to-contaminant ratio is desired for isotope measurements.

Alternatively a proton-to-neutron converter setup can be utilised for subduing contaminant species, whereby the GeV protons are instead directed onto a solid tungsten rod situated adjacent to the target unit [Cat03; Lui12; Got14]. With spallation as the predominant reaction channel, neutrons are produced with $\sim$ MeV kinetic energies which are subsequently used to induce fission within the target material. Not only is this particularly effective for purifying neutron-rich isotope beams by suppressing the production of isobaric contaminants from other nuclear reaction mechanisms, it also increases the longevity of the target unit because of a reduced beam power deposition. Its current geometry however, has a poor solid angle coverage with respect to the emitted neutrons and ultimately results in a reduced beam intensity. New designs have already been commissioned with the aim of eliminating this detriment, in addition to further reducing the proton flux seen by the target material [Got14; Ram19a]. With respect to neutron-rich gallium isotope beams at ISOLDE, the geometry

proposed in Ref. [Got14] could potentially suppress prevalent ^{37}Rb and ^{38}Sr isobaric backgrounds by up to a factor of 5, while preserving the gallium isotope yields obtained with direct proton bombardment [Ram19b].

Predetermining the proton-pulse sequence should also be considered for maximising the experimental efficiency, however considering the demand has to be shared with other experimental facilities at CERN, in reality this mostly cannot be done. This does not pose much of a problem for isotopes with high production yields, whereby measurements can be performed by typically spending $\sim 1\text{--}2$ s on each frequency step. It is more crucial for those with much lower yields however, otherwise fluctuations in the observed yield become more pronounced and can affect the interpretation of observed hyperfine structure. While the CRIS setup does not rely on any proton-related triggering signals from either CERN or ISOLDE during an experimental campaign and is thus, ‘freely running’, temporal data regarding proton-pulse impacts with the target are usually recorded during any hyperfine-spectroscopy measurements. As such, it is possible to synchronise with respect to the proton pulses or supercycle post-experiment in an offline analysis. The latter remedy had been utilised for the case of ^{214}gFr in Chapter 7.1.2, in which measurements relied on spending two full proton supercycles (each containing 30 proton pulses, resulting in a total time of 72 s) for each frequency step.

3.1.2 Isotope ionisation

After leaving the target material, isotope products travel through a transfer line attached to the target setup. If a transfer line is made from a material with a high work function (such as tantalum or rhenium) and also heated in excess of 2000 K, the surface ionisation process is enhanced. Such ion sources are sufficient for ionising elements with low ionisation potentials. Further yield enhancements can be done with the Resonance Ionisation Laser Ion Source (RILIS) [Mar13; Rot13] and is utilised by the majority of experimental setups at ISOLDE. Using a specific atomic transition, lasers are used to selectively ionise the isotope of interest through step-wise excitations. Although it can provide higher degrees of selectivity and efficiency, it still does not effectively deal with isobaric species that can be easily surface ionised. New ion-source developments, such as the Laser Ion Source Trap (LIST) [Sch10; Fin15] and the Versatile Arc Discharge and Laser Ion Source (VADLIS) [Day17; Mar18b; Mar19], are currently under investigation in an effort to further reduce this type of contamination.

3.1.3 Isotope acceleration and separation

Newly-ionised radioactive species are subsequently accelerated by a voltage of up to 60 kV; for CRIS, this is normally in the range of 30-40 kV. Travelling along the direction of the third arrow in Figure 3.1, at the end they reach the High-Resolution Separator (HRS) where under the application of a homogeneous magnetic field, B , the ion trajectories are bent along a circular path characterised with a radius, R . Under these conditions, equating the magnetic force exerted by an ion with mass m , velocity v , and charge q , with its centripetal force yields:

$$Bqv = m \frac{v^2}{R} \quad \rightarrow \quad B = \frac{1}{R} \sqrt{\frac{2mV}{q}}, \quad (3.1)$$

with an additional substitution concerning the kinetic energy gained by accelerating an ion through a voltage V : $mv^2/2 = qV$. The succinct consequence from (3.1) states that a particular isotope with mass m can be isolated from the other masses just by using a precise B field. Two magnets comprise the HRS system, each with a bending radius of 1 m and steering the beam through 90° and 60° respectively. This system combined has a high mass-resolving power of approximately $m/\delta m = 6000$ thanks to elaborate ion-optical systems [Cat17], however it should be stressed that this is still not sufficient to filter out all isobaric contamination.

Another separator system is also available at ISOLDE: the General-Purpose Separator (GPS). It is used if the isotope beam is obtained using the other target station mentioned in Chapter 3.1.1. Consisting of just a sole magnet with a bending radius and angle of 1.5 m and 70° respectively, the available mass resolution available is lower than the HRS with $m/\delta m \approx 800$. However its advantage is the ability to allow for the simultaneous extraction of up to three mass separated beams with a mass spread of up to $\pm 13\%$ around the primary beam [Cat17].

3.1.4 Bunched beams with the ISCOOL

Applicable to all CRIS experiments at ISOLDE, the use of the HRS system has the added advantage of being able to bunch the beam using the Isolde COOLER ISCOOL: a radio-frequency quadrupole cooler-buncher [Frå08; Man09b], which is mounted just behind. Ions entering ISCOOL are slowed down through collisions with a buffer gas contained within, surrounded by electrostatic quadrupole elements. Their plates are held at voltages consisting of a constant direct-

current component to facilitate the drifting of ions towards the end of ISCOOL, and an oscillating component which combined together traps these ions in this region. The end plate of this setup constantly switches between +60 V and 0 V within a ns timeframe, resulting in a succession of ion bunch accumulations (up to $\sim$ ms at a time) and releases with a typical temporal width of 1-5 μ s and a repetition rate of the order of 100 Hz. ISCOOL transmission efficiencies can reach up to 80% for the heavier isotopes, however this dramatically reduces for the lighter species because of radio-frequency heating losses within the buffer gas [Fr 08].

It is incredibly advantageous to work with bunched beams at CRIS because it allows them to be temporally overlapped with laser pulses from two (or more) laser beams used for the resonance ionisation, minimising laser duty cycle losses and maximising the experimental efficiency; see Chapter 3.1.2 for further details. Possible short-lived ($\sim$ ms) isomeric states in the ion beam can also be crudely inferred by measuring with ISCOOL accumulation times spanning several half-lives, which in theory should significantly suppress its presence with respect to the longer-lived ground state in any hyperfine spectrum. Especially if its appearance is unexpected and amongst background of comparable intensity, this procedure could help to distinguish between its possible existence from a statistical fluctuation. Subsequent justification for scanning additional frequency ranges in order to find the rest of its structure could also result, if time allows during an experimental campaign. A dedicated CRIS cooler-buncher is also currently under development [Coo19], which would allow for bunched beams at the CRIS setup not only in conjunction with the GPS, but also during offline characterisation tests.

After passing through the ISCOOL, the ion bunches are re-accelerated back to their previous voltage (of up to 60 kV) and can then be sent to the CRIS setup for laser spectroscopy measurements.

3.2 Collinear Resonance Ionization Spectroscopy

As mentioned before in Chapter 2.3, laser spectroscopy principles are by no means new concepts. Although the CRIS experiment is relatively new at the ISOLDE facility [Fla13a], it relies on the merger of two very well established laser spectroscopy techniques: collinear laser spectroscopy, and resonance ionisation spectroscopy. Acting as an interlude before outlining the procedure of measuring the hyperfine structure of isotope beams at the CRIS setup, the details of the constituent laser techniques are first outlined.

3.2.1 Collinear Laser Spectroscopy

Collinear Laser Spectroscopy (CLS) involves the spatial overlapping of continuous-wave high-resolution laser light with an accelerated isotope beam. Already well established at the ISOLDE facility for nearly 40 years [Neu17], its key advantage lies in the high-resolution measurements that can be performed. Older optical-spectroscopy setups, such as an electric discharge lamp and Fabry-Pérot interferometer combination, are severely limited by the Doppler broadening induced by the thermal energy of the atoms. At a temperature T , the frequency width $\delta\nu_D$ is characterised by a Maxwellian velocity distribution:

$$\begin{aligned}\delta\nu_D &= \nu_0 \sqrt{\frac{8\ln 2 kT}{mc^2}} \\ &\approx \frac{214}{\lambda_0} \sqrt{\frac{T}{m}} \text{ GHz},\end{aligned}\tag{3.2}$$

where λ_0 represents the transition wavelength (in nm) used to probe an isotope with atomic mass³ m . Its shape is Gaussian in nature and for ^{214}Fr the linewidth amounts to $\delta\nu_D \sim 0.6$ GHz at room temperature. Especially for the lighter species, typical GHz widths are usually sufficient to encompass entire hyperfine structures, thus making it impractical to measure them. The defining aspect of CLS developments designed to overcome this limitation involves working with an accelerated ion beam.

While each ion gains the same kinetic energy increase, their energy spread δE remains invariant with respect to their initial spread as they exit the target:

$$\delta E = \delta\left(\frac{1}{2}mv^2\right) = mv\delta v.\tag{3.3}$$

The consequence of (3.3) is that reduced velocity spreads δv can be achieved with higher acceleration voltages V , according to $v = \sqrt{2eV/m}$. Additionally considering the definition of the Doppler shifted width $\delta\nu_D = \nu_0\delta\beta = \nu_0(\delta v/c)$, their insertions into (3.3) yields for the Doppler width:

³At the time of writing, Ref. [Wan17] provides an up-to-date directory for atomic masses of all observable isotopes.

$$\begin{aligned}
\delta\nu_D &= \nu_0 \frac{\delta E}{\sqrt{2eVmc^2}} \\
&\approx \frac{6921}{\lambda_0} \sqrt{\frac{\delta E}{mV}} \text{ GHz},
\end{aligned}
\tag{3.4}$$

with δE expressed in units of eV. At the ISOLDE facility, typical accelerated energy uncertainties of $\delta E \sim 5$ eV are common [Cat17]. Using ^{214}Fr once again as an example, accelerating a beam to $V = 40$ kV results in a Doppler width of $\delta\nu_D \sim 12.5$ MHz. In contrast, the natural linewidth $\delta\nu_n$ represents a sort of theoretical limit to the resolution that can be obtained for a particular transition. Related to the mean lifetime τ of the excited atomic or ionic state via the Heisenberg uncertainty principle:

$$\delta\nu_n = \frac{1}{2\pi\tau}, \tag{3.5}$$

which is characteristically Lorentzian in nature. For the 422.7-nm transition in neutral francium involving the excited $8p\ ^2P_{3/2}$ state with an 83.5(15) ns lifetime [Aub04], this amounts to $\delta\nu_n \sim 2$ MHz. Therefore, CLS on an accelerated beam represents an impressive orders-of-magnitude reduction in the lineshape broadening towards on-par with the natural linewidth of the transition, provided a sufficient acceleration voltage is chosen. Power-broadening effects however, can increase the width given in (3.5), according to:

$$\delta\nu_P = \delta\nu_n \sqrt{1 + \frac{I}{I_s}}, \tag{3.6}$$

where I represents the laser intensity and I_s denotes the intensity at which the transition under study becomes saturated. Using more-intense laser powers usually increases the rate of stimulated emission; the subsequent reduced transition lifetime broadens the natural linewidth according to (3.5). While power broadening manifests to some degree in most circumstances, it is usually beneficial to choose a laser power below I_s when performing any isotope measurements, in order to minimise these effects. The determination of I_s can be done by observing the obtained count rate for a range of laser-power values; an analysis of such tests for the 417.2-nm transition in neutral gallium can be found in Chapter 3.3.2.

It is important to remember that accelerated ions observe a laser frequency ν which is Doppler shifted with respect to the laboratory frame of reference ν_L :

$$\nu = \nu_L \frac{1 \pm \beta}{\sqrt{1 - \beta^2}}, \quad (3.7)$$

for collinear (-) or anti-collinear (+) geometries. β represents the velocity ratio⁴ of the ions:

$$\beta = \frac{v}{c} = \sqrt{1 - \left(1 + \frac{eV}{mc^2}\right)^{-2}}, \quad \text{or} \quad = \sqrt{\frac{2eV}{mc^2}}, \quad (3.8)$$

with respect to the speed of light c . Ion velocities observed at the ISOLDE facility are non-relativistic; using $V = 30$ kV, ^{65}Ga and ^{214}Fr ions only travel at $v \sim 0.0010 c$ and $0.0006 c$ respectively. Excitation of the atom occurs whenever the frequency in (3.7) matches that of a hyperfine transition, which subsequently decays either back to the ground-state or some other low-lying state while emitting fluorescence. Diagram (a) of Figure 3.2 depicts a representative transition scheme for these processes. PhotoMultiplier Tubes (PMT) are usually used to detect this fluorescence and are placed as close as possible to the path of decaying species in order to maximise the solid-angle coverage. Despite typical 10% - 40% detection efficiencies of PMT, scattered laser can significantly contribute to the overall background and means that measurements are usually limited to isotopes produced with yields of $\sim 10^3 \text{ s}^{-1}$. Special cases however have allowed this limit to reach $\lesssim 10^2 \text{ s}^{-1}$, such as: gating on specific β -particle [Mar11] or γ -ray [Fla12] transitions with the ion/atom bunch, and experimental setup improvements to reduce observed background limits [Gar16].

Obtaining hyperfine spectra using CLS can be done by scanning with respect to either β in (3.8), or ν_L in (3.7). The former method relies on scanning the acceleration voltage V while locking the laser to a particular value of ν_L ; V can be accurately defined and quickly scanned, so measurements can be performed quickly and repeatedly with ease. Alternatively, scanning with ν_L has the advantage of being able to scan over vast frequency ranges > 30 GHz in one attempt. Nonetheless, a well-characterised laser system is imperative because otherwise, any laser-power, -pointing, or -mode instabilities would easily manifest

⁴Both expressions are valid; with respect to its use in (3.7), the difference between the two amounts to < 0.2 MHz for all available masses and acceleration voltages at the ISOLDE facility.

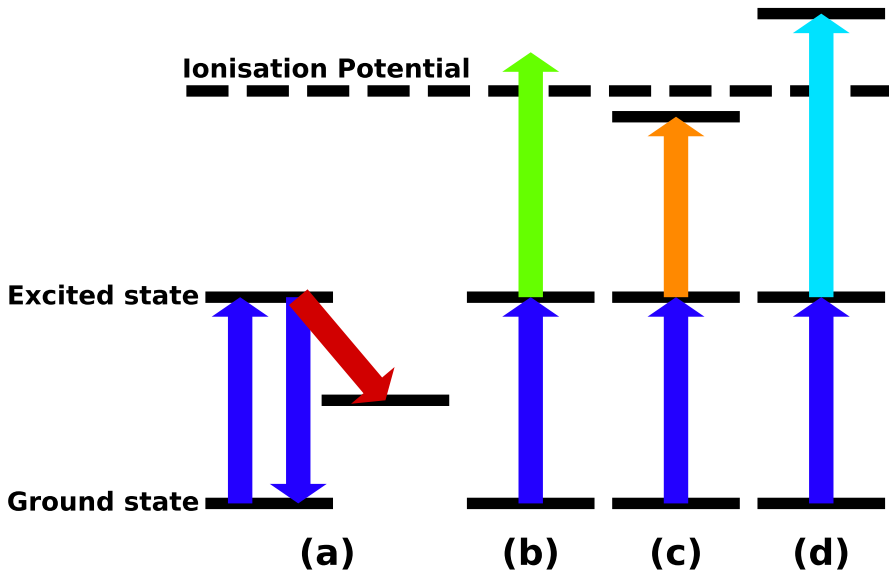


Figure 3.2: A selection of various transition schemes available for laser spectroscopy. Starting from a transition to an excited state, its hyperfine structure can be probed by detecting either (a) the decay to another state, or ions created: (b) through complete ionisation, (c) by exciting to a high-lying Rydberg state, or (d) by exciting to an auto-ionising state.

within any isotope measurement; those involving lighter species with very small hyperfine splittings would be most detrimentally affected. At the CRIS setup for example, the laser setup utilised during the 2017 IS620 experiment measuring ^{19}K isotopes [Yan16a] required an exceptional stability, such that the associated hyperfine parameters and isotope shifts could be extracted with a ≤ 1 MHz precision [Kos19b].

3.2.2 Resonance Ionisation Spectroscopy

Resonance Ionisation Spectroscopy (RIS) relies on detecting ionised species resulting from the step-wise excitation and subsequent ionisation of an atomic system using two or more lasers. Additional diagrams displayed in Figure 3.2 show possible routes for the production of ionic species. The most direct method uses a non-resonant transition to ionise past the Ionisation Potential (IP) in diagram (b), typically with either 1064-nm or very intense 532-nm light.

Alternatively, excitation to either a high-lying Rydberg state in diagram (c) or an auto-ionising state in diagram (d) can be performed, provided they exist and their cross sections are favourable. The latter scenario normally requires less-intense laser powers, thus reducing the probability of ionising isobars if 532-nm light is used for complete ionisation.

Particle detectors such as a Micro-Channel Plate (MCP) or a secondary electron multiplier can be used for the detection of ions, with a much more favourable detection efficiency: close to 100%, in comparison to the $\leq 40\%$ offered by using CLS techniques. Additionally, background-related events with RIS originate from different sources to that of the signal (*e.g.* from collisional ionisation processes), whereas both the signal and background in CLS methods are related directly to the laser beam. As a consequence, measurements of isotopes with yields of a few tens per second can be routinely made [Gro17b].

High degrees of selectivity can also be obtained as a result of the laser-atom interaction. Denoted S , it is defined as the probability of exciting a selected isotope with respect to that of a neighbouring isotope. For a multi-step ionisation process, in which Δ represents the frequency difference between two neighbouring elements or states and Γ denotes the interaction linewidth, it can be numerically evaluated by [Cam16]:

$$S \approx \sum_{n=1}^N 4 \left(\frac{\Delta}{\Gamma} \right)^2 \approx \sum_{n=1}^N S_n, \quad \text{for } \Delta \gg \Gamma, \quad (3.9)$$

where S_n represents the selectivity of an individual transition. Thus, it is easy to imagine how a high selectivity can be obtained, thus potentially allowing for decay-spectroscopy [Lyn13; Lyn14] and mass [Van02] measurements to be performed on purely isomeric beams. If particle detection is replaced with tagging to specific decay radiation, the selectivity is further enhanced; this has allowed for successful isotope measurements with yields of $\leq 0.1 \text{ s}^{-1}$ [Sel13; Laa16; Sel19].

3.3 The CRIS setup at ISOLDE

Combining the pinnacle features of CLS and RIS in Chapters 3.2.1 and 3.2.2 together, CRIS is able to measure the hyperfine spectra of isotopes with high resolutions, efficiencies, selectivities, and background suppressions, even if these isotopes are scarcely produced. A schematic of the experimental setup is shown

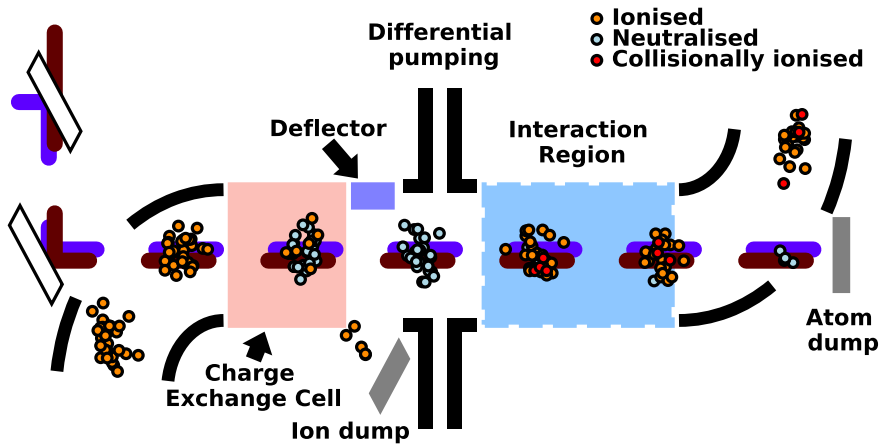


Figure 3.3: A schematic of the CRIS experimental setup at ISOLDE; dimensions are not to scale. A colour code for the different charged states of a particular species is given in the upper-right corner. See text for further descriptive details regarding each important beamline section.

in Figure 3.3 and this next section continues the journey of the bunched-ion beams after the end of the red arrows in Figure 3.1.

3.3.1 Neutralisation with the charge-exchange cell

Upon receiving bunched-ion beams from the ISCOOL at ISOLDE, they are first focused using a pair of quadrupole-triplet elements. After subsequent electrostatic deflection through a 30° bend, they then enter into a Charge-Exchange Cell (CEC). If an atomic⁵ transition is being probed, the CEC is filled with an alkali vapour with the aim of neutralising the ion bunches by collisional transfers of charge (in Figure 3.3: orange to blue, from the second to the third bunch). Upon exit towards the differential-pumping region, any remaining non-neutralised ions are deflected away from the main beam.

In general, neutralisation efficiencies and choice of alkali vapour are bespoke for each element under study [Ver19b] and neutral fractions of between 50%-70% are common. While this had been achieved for francium isotopes throughout

⁵For probing ionic transition schemes, the ion bunches would just pass through an empty CEC with no subsequent neutralisation. For some elements however, this represents an unfavourable choice because of the deep Ultra-Violet (UV) light required to probe such transitions, which can be more difficult to produce with a sufficient intensity [Neu17].

the IS471 experiment in 2014 using a potassium vapour [Gro15; Lyn16; Far17], a maximum of $\sim 25\%$ could only be obtained for gallium isotopes using the same vapour during the IS571 experiment in 2015 [Far17]. An efficiency of up to 70% has been reported by instead choosing sodium vapour [Pro13] and has already been attempted during the 2017 IS571 experiment. Unfortunately, this could not be confirmed at the CRIS setup because of Faraday-Cup (FC) failures throughout the latter experiment. Thus, the conduction of neutralisation tests before an experiment is imperative to ensure the appropriate selection of neutralisation vapour.

3.3.2 Laser-ionisation schemes in the interaction region

After passage through the CEC and differential-pumping sections, the atom bunches enter an Ultra-High Vacuum (UHV) Interaction Region (IR) in which the process of CRIS happens (in Figure 3.3: blue to orange, from the fourth to the fifth bunch). Incredibly low pressures are required to suppress collisional re-ionisation processes from any gas molecules present (in Figure 3.3: blue to red, from the fourth to the fifth bunch), representing a majority of the produced background when using RIS techniques. Pressures of 9.0×10^{-9} mbar, 1.6×10^{-8} mbar, and 2.3×10^{-9} mbar have been achieved for the 2014 IS471 francium, 2015 IS571 gallium, and 2017 IS571 gallium experiments respectively, however typical pressures can now regularly reach towards the lower-end of 10^{-10} mbar [Kos19a]. The differential pumping section not only is responsible for these low pressures considering the proximity of the CEC (allowing for a pressure differential of up to 10^{-3} mbar), but also prevents alkali vapour from entering the UHV region which would otherwise destabilise the low pressure. Atomic bunches arriving inside the UHV are spatially and collinearly overlapped with the laser light, with a precise time synchronisation in order to minimise duty cycle losses and subsequently maximise the experimental efficiency. Laser-setup requirements for the francium and gallium transition schemes used at the CRIS setup will now be detailed in the next two sections.

N.B. Wavelengths quoted to different precisions (for example: 422 nm, 422.7 nm, and 422.69 nm) are interchangeable not only in these sections, but also throughout the rest of this thesis.

Francium

The ionisation scheme used for the study of francium is shown in Figure 3.4. It is a two-step scheme: an initial 422.69-nm step to excite francium atoms from the $7s\ ^2S_{1/2}$ ground state to the $8p\ ^2P_{3/2}$ excited state at 23658.306 cm^{-1}

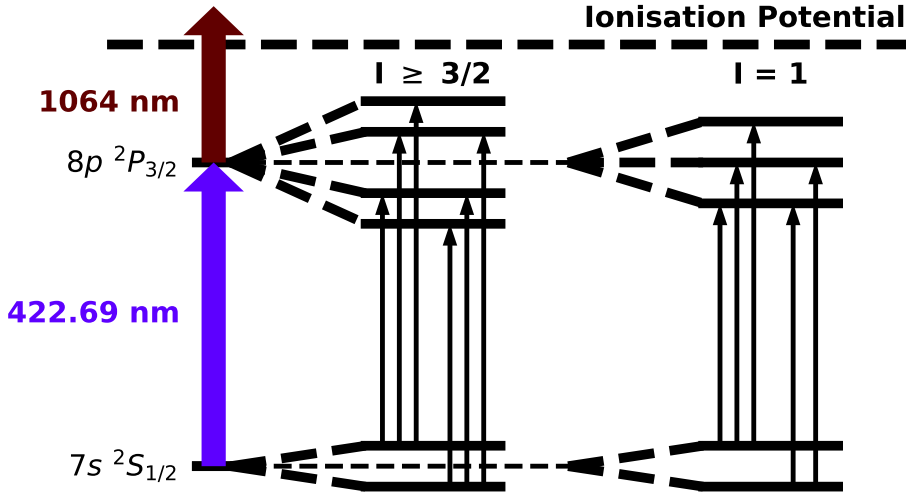


Figure 3.4: The two-step resonance ionisation scheme used to study neutral francium at the CRIS setup. Allowed transitions for $I \geq 3/2$ and $I = 1$ spin assignments are also given.

(also known as the $D'2$ transition), followed by a non-resonant 1064-nm step for complete ionisation past the IP at 32848.872 cm^{-1} [San07]. This transition, first studied in 1987 [Duo87], has been utilised in every francium study at CRIS [Fla13b; Lyn14; Bud14; Gro15; Lyn16; Far16; Wil17b]. Its spin-orbit partner $8p\ ^2P_{1/2}$ has also been studied using 432.54-nm light [Duo87]. Additionally, transitions to the $7p$ level have been explored, utilising 717.99-nm ($D2$) [Coc85; San09; Vos13; Vos15] and 816.94-nm ($D1$) light [Col14] for the associated $^2P_{3/2}$ and $^2P_{1/2}$ states respectively.

422.69-nm light has been produced in two ways:

- Narrowband light was obtained by frequency-doubling 2.7 W 844-nm fundamental light from a Matisse TS continuous-wave Titanium:Sapphire (Ti:Sa) laser pumped by a Coherent Verdi G18 15 W laser, using a Wavetrain external cavity with a 10% efficiency. Short pulses were produced by switching the laser-light polarisation using a Pockels cell and polarising beam-splitter cube setup; the applied voltage on the Pockels cell rapidly switched between 0 and +2.4 kV with the use of a Behlke FSWP91-01 fast-square wave pulser. Further information on this system is given in Ref. [Gro15].

- Broadband light was provided by the RILIS team: frequency-doubled 844 nm light from a tunable Ti:Sa laser pumped by the second harmonic output of a Photonics Industries DM-60-532 Nd:YAG laser operating at 10 kHz, using a BBO crystal. Fibre-coupling was used to transport the light to the optical tables at CRIS, using a 35-m Thorlabs SFS105/125Y optical fibre. Further information on this system is given in Ref. [Rot13].

Although the broadband setup had been used previously for commissioning tests and initial experimental campaigns at the CRIS setup [Coc13b; Fla13b; Lyn14], it was utilised during the end of the francium experiment in 2014 because of a failure of the CRIS narrowband laser setup. This explains the differing resolutions of ^{221}Fr measurements observed in Chapter 7.1.1.

Fundamental 1064-nm light was produced using a Litron LPY 601 50-100 PIV Nd:YAG laser system, consisting of two laser heads each capable of delivering 80 mJ at 100 Hz. These heads could either be operated independently, or combined together for a maximum repetition rate of 200 Hz; this represents an improvement from the 30 Hz duty cycle of an older laser system CRIS measurements [Fla13b]. With 200 Hz able to match a cooling and bunching period limit of 5 ms, measurements of short-lived isotopes close to the limit of ISOL production can be performed with minimal decay-related yield losses. This has proved pivotal for the successful measurements of ^{214g}Fr with a half-life of only 5 ms, which can be found in Chapter 7.1.2.

Gallium

In contrast to francium, the gallium ionisation scheme consisted of three parts: a 417.20-nm step to excite gallium atoms from the $4s^24p\ ^2P_{3/2}$ metastable state at 826.190 cm^{-1} to the $4s^25s\ ^2S_{1/2}$ excited state at 24788.530 cm^{-1} and probe the hyperfine structure, a 639.66-nm step to further excite to the $4s^26p\ ^2P_{3/2}$ excited state at 40417.620 cm^{-1} , and a subsequent 1064-nm non-resonant step for complete ionisation past the IP at 48387.634 cm^{-1} [Shi07]. It represents the first three-step scheme used to probe hyperfine structure at the CRIS setup. The 417.20-nm transition in gallium has already been studied at the ISOLDE facility [Che10b; Che10a; Man11a; Pro12; Che12a], with selected isotopes also measured using the 403.30-nm transition which starts with the $4s^24p\ ^2P_{1/2}$ ground state.

417.20-nm light was produced in different ways for the two gallium experimental campaigns at CRIS:

- The first IS571 experiment in September 2015 used frequency-doubled 834-nm light from an M-Squared SolsTiS⁶ continuous-wave Ti:Sa laser, using an ECD-X frequency doubler. It was pumped with 18W of frequency-doubled 532-nm light from a Lighthouse Photonics Sprout G-18W Nd:YAG diode-pumped solid-state laser; short pulses were created by utilising the same Behlke setup which produced narrowband 422.69-nm light for 2014 IS471 francium experiment. Immediately after the doubler, a maximum of 2.5 W of 417.20-nm light could be produced.
- The second IS571 experiment in October 2017 used frequency-doubled light from an injection-seeded Ti:Sa⁷ laser, using a BBO crystal. 1 mW of fundamental 834-nm light from the SolsTiS in the previous setup was used as the seed, pumped with 1.2W of frequency-doubled 532-nm light from a Lee Laser LDP-100MQ Nd:YAG laser operating at a 1 kHz repetition rate. After being doubled, a maximum pulse energy of 60 μJ with a temporal width of 50 ns could be produced.

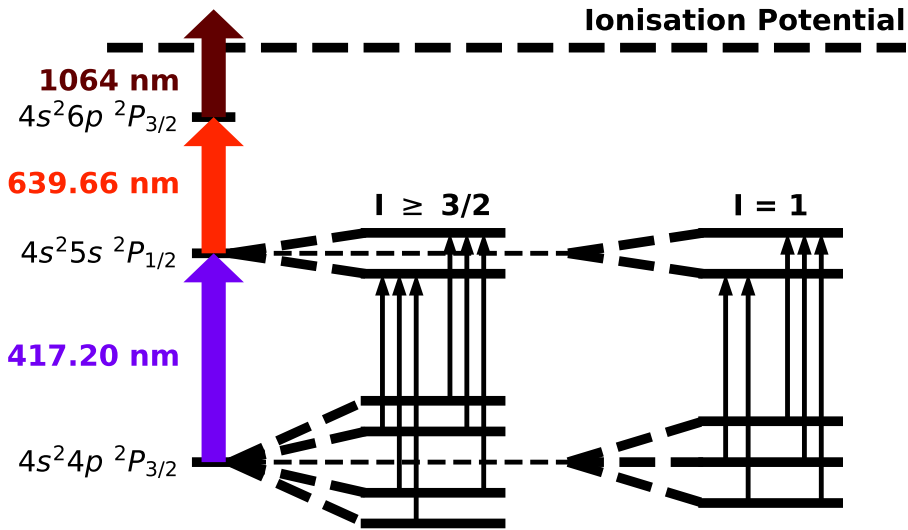


Figure 3.5: The three-step resonance ionisation scheme used to study neutral gallium atoms at the CRIS setup. Allowed transitions for $I \geq 3/2$ and $I = 1$ spin assignments are also given.

⁶Its range spans 720-970 nm with a peak power output of 6 W at 780 nm.

⁷This laser system was developed in collaboration between the Johannes Gutenberg-Universität Mainz and the University of Jyväskylä; see Refs. [Kes08; Son17] for further information.

Light required for the other two steps was produced from the same laser setups for both IS571 experiments. Fundamental 639.66-nm light was obtained from a Spectron Spectrolase 4000 pulsed dye laser, using DCM dye with concentrations of 0.3 g/L and 0.044 g/L in the resonator and amplifier respectively. It was pumped with 50 mJ of frequency-doubled 532-nm light from one of the Litron laser heads, using a half-wave plate placed before the second-harmonic generation crystal. With an approximate linewidth of 10 GHz, output pulses could be produced with a maximum energy of 4 mJ. The other Litron laser head provided the 1064-nm light for the ionisation step; newly installed Litron flashlamps for both experiments meant pulse energies of up to 260 mJ could be produced.

Saturation tests have also been conducted for all three steps of this transition scheme, during the first IS571 experiment. A saturation value P_0 can be found by considering the following relationship between the count rate I and the power or energy of the laser pulse P [Dem15]:

$$I(P) = A \frac{P/P_0}{1 + P/P_0}. \quad (3.10)$$

Available data and corresponding fits from (3.10) are given in Figure 3.6. Although saturation for the 417-nm and 639-nm transitions could quite easily be

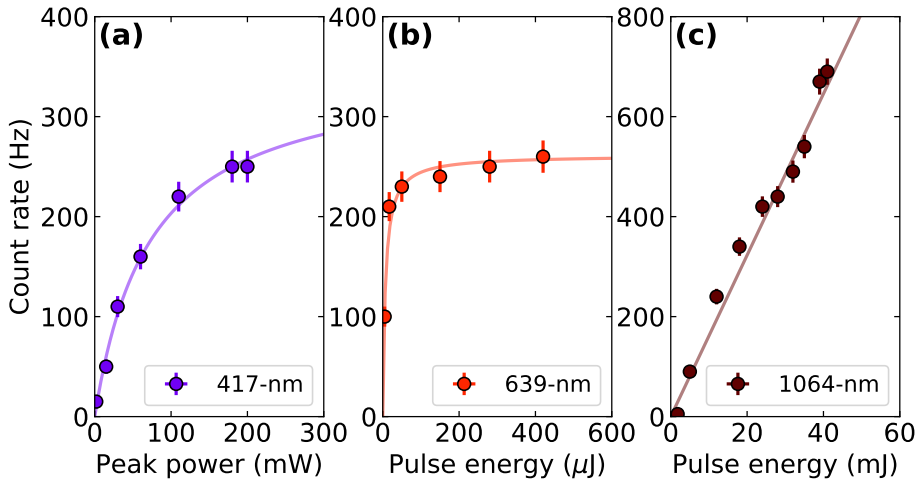


Figure 3.6: Saturation curves for the: (a) 417-nm and (b) 639-nm resonant, and (c) 1064-nm non-resonant transitions in Figure 3.5. Data have been taken from the run-logbook used in the 2015 IS571 gallium experiment.

achieved with 72(12) mW and 7(1) μ J respectively, the absence of any asymptotic behaviour with the 1064-nm light indicated that this could not be achieved for the final non-resonant ionisation step, even at the highest pulse energies available. While using the highest powers available may seem advantageous for improving the overall ionisation efficiency and thus, the strength that an isotope measurement appears in a hyperfine spectrum, adverse atomic effects may manifest as a result. For this gallium transition scheme, the use of such high-power laser light resulted in the occurrence of lineshape distortions for all observable resonances; Chapters 4.4.2 and 4.4.3 provide further insights, characterisations, and discussions into this phenomenon.

3.3.3 The detection of hyperfine structure

Newly re-ionised ion bunches exit the UHV IR and are subsequently deflected through a 20° bend. The outer plate has a hole allowing for non-ionised atoms and laser light to travel towards an atom/beam dump. For the experimental campaigns described in this thesis, ion detection involved two Hamamatsu F4655-12 MCP detector setups past this electrostatic element. Initial detection utilised an MCPe located 1.8 m away from the 20° bend inside a Decay Spectroscopy Setup (DSS). Off-axis with respect to the beam path and normally biased up to +2.4 kV, it detected emissions of secondary electrons created by steering the ions onto a negatively charged copper dynode. Hyperfine structures obtained during the IS471 francium experiment in 2014 relied on this setup. Complementary α -decay spectroscopy studies were also performed inside the DSS setup, whose description is detailed in Chapter 3.3.4.

A poor and broadened ion-beam transmission resulting from the geometrical nature of the 20° bend presented a major hindrance when using the MCPe for ion detection. Efforts to enhance the overall experimental efficiency motivated the installation of the MCPi detector, located only 53 cm away from the 20° bend. Biased at -2.4 kV, it directly detects the incoming positively-charged ions. It is mounted on a pneumatic linear drive which allows it to be moved out of the ion beam path if any decay spectroscopy study needs to be performed at the DSS. Hyperfine structures obtained from both IS571 gallium experiments in 2015 and 2017 primarily used this setup, however the former MCPe detector was sparingly used, only for whenever its successor suffered from voltage supply failures. Additional details for both MCP setups used at the CRIS experiment can be found in Ref. [Gro17c]. Latest developments have replaced the MCPi detector with an ETP DM291 MagneTOFTM detector [Ver19b], providing further sensitivity for the lowest ion count rates. Microamp beams can also be measured, which would otherwise quickly render a standard MCP detector inoperable.

3.3.4 Decay-spectroscopy studies at the CRIS setup

Complementary decay spectroscopy studies can also be performed on the isotope beam using the DSS at the CRIS setup. Alpha-decay spectroscopy has already been performed on ^{202}Fr , ^{204}Fr , ^{206}Fr , and ^{214}Fr : the first three as a means for assigning hyperfine transitions to a particular state [Lyn14; Lyn16], and the latter for spin assignment arguments, as presented in Chapter 7.2.3.

Beam is provided to the DSS by removing not only the MCPi/MagneTOF from the beam path, but also any voltage on the copper dynode. Biasing the electrostatic steering plates can additionally be done, in helping to direct the beam to the DSS. Figure 3.7 shows schematics for the DSS1.2 setup for α -decay spectroscopy. Ions first pass through a Canberra ANPD300-18-300RM Annular Passivated Implanted Planar Silicon (APIPS) detector with a 4 mm aperture, and impinge on one of eight available carbon foils. A collimator protects the back from direct implantation and is electrically insulated so that beam intensities can be measured during any ion-beam transport optimisations. A Canberra A300-17AM PIPS detector is situated behind the implantation foil. Both detectors feature a Si thickness of $300\text{ }\mu\text{m}$, suitable for α and β charged-particle detection at the DSS. The carbon foils are part of a rotatable wheel system holding up to a maximum of ten, each with a $20\pm 1\text{ }\mu\text{g}/\text{cm}^2$ thickness (produced at the GSI target laboratory [Lom02]). Two of these positions are reserved for: a copper disk which acts as a FC for beam-tuning measurements, and two $50\text{ Bq }^{241}\text{Am}$ sources positioned back-to-back for calibrating the APIPS and PIPS detectors. Analogous to Mylar tape-transport setups, this system addresses the removal of long-lived contamination while also offering a more compact setup. A stepper motor is also available for the remote rotation of the wheel; manual rotation is also possible for longer collection times. Supplementary DSS information and characterisations are detailed in Refs. [Raj13; Lyn13].

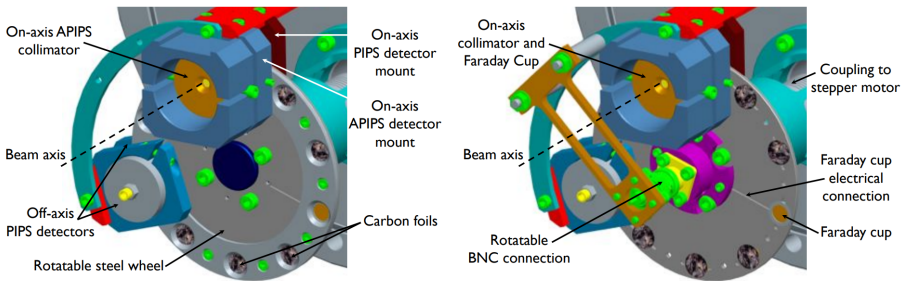


Figure 3.7: Schematic drawings of the DSS1.2 setup at the CRIS setup. Figures are taken from Ref. [Lyn13].

Another alternative setup is the DSS2.0 [Lyn17], which has been developed as a versatile upgrade to its DSS1.2 predecessor; further details can be found in the aforementioned reference. With a similar silicon-detector configuration, carbon foils are instead attached to a movable linear actuator. Although it is only limited to two carbon foils and a Faraday cup, its aluminium construction with a 10 cm cross-section and thin 2 mm braised aluminium walls, provides a better γ -ray detection efficiency because of the reduced attenuation and greater solid angle that can be subtended. Additionally, its compact nature also allows the possibility of incorporating a tape transport system. So far, it has already been used to analyse the beam content of pure-state polonium beams [Alt17].

The recent installation of plastic scintillators around the outside of the DSS1.2 has allowed for β -decay assisted laser spectroscopy at CRIS. Hyperfine spectra are obtained by observing the β -decay rate of isotopes impinged onto an aluminium flange installed at the end of the beamline, while scanning the laser frequency. It can be advantageous for measuring isotopes which suffer from excessive levels of stable isobaric contamination; provided a sizeable distinction in the β endpoint energies, such background can be suppressed by applying a large threshold value on the scintillators or using an appropriate attenuator. At present, hyperfine-structure measurements of ^{52}K have successfully been obtained using this experimental technique [Kos19a].

3.4 Summary

At the forefront of isotope beam production, the ISOLDE facility is home to an assortment of experimental setups which have provided some of the most exciting discoveries in the nuclear physics community. One of these setups is the CRIS experiment: employing both CLS and RIS techniques, the hyperfine structures of exotic nuclei can be easily measured with high resolutions and efficiencies. With additional decay-spectroscopy facilities available to help further aid these measurements, the CRIS setup is well set for probing some of the most puzzling nuclear-structure phenomena across the nuclide chart.

Chapter 4

Analytical tools for the interpretation of hyperfine structure

While the CRIS experiment represents a rather impressive feat in the efficient detection of exotic isotopes, the experimental details are only part of the ingredients required for potentially explaining nuclear-structure phenomena with success. A rigorous analytical framework is needed for extracting the hyperfine observables from the resulting raw data with confidence. The following chapter will discuss the procedure for raw data collected at the CRIS setup, alongside appropriate models for their interpretation.

4.1 Summarising CRIS-setup measurements

This section summarises the gallium and francium measurements performed at the CRIS setup, the analyses of which feature later in this thesis in Chapters 5, 6, and 7. Where applicable, experimental efficiencies and yield estimates are stated for a given experimental campaign.

4.1.1 For gallium isotopes

Table 4.1 gives an overview of the gallium laser-spectroscopy measurements performed throughout the two IS571 (in 2015 and 2017) experiments at the CRIS setup. Scan numbers correspond to an identifier present in the names of associated raw-data files, with Chapter 4.2 further detailing their file structures. Each represents a full hyperfine-spectrum measurement; those enclosed in square brackets represent a combined data set in which an analysis has been performed on. Although no discernible resonances have been observed for $^{62,83}\text{Ga}$, their measurements are nonetheless included in this table for reference and are further discussed in Chapters 8.1.1 and 8.1.2.

Table 4.1: A summary of gallium isotope measurements performed at the CRIS setup. The total time spent collecting data for each isotope is displayed to the right; target-material information is provided in the footnotes below.

A	2015 IS571: Target No.548 ¹		2017 IS571: Target No.595 ²	
	Scan #	Time	Scan #	Time
62			[45,47,48,55]	08:04:29
65	128, 129	00:35:26	44	00:13:53
67	76, [99,100], 127	01:23:05		
69	13, 14, 15	01:17:15		
71	16, 17, 26, 54, 55, 62, 72, 78, 85, 88, 98, 101, 102, 103, 116, 117, 125, 131	06:36:56	36, 43, 51, 53, 57	01:57:08
75	28, 31	01:30:41		
79	[32,33], [63,66]	01:15:58		
80	90, 91, 96	02:30:30	35, 58, 59	02:05:22
81	70, 71	00:44:28		
82	[73,74,75,86,87, 105,118,120,123,124] ,[106,107,108,109,111,121,122]	18:36:34		
83	[56,57,58], [59,60,61]	03:43:05		
		38:13:58	12:20:52	

¹A UC_x-Ta target with a neutron-converter setup, installed on Sept. 3rd for IS571 lasting Sept. 22th - 30th. This target contained Mg, Na, and Ga mass markers; previous irradiation of the neutron converter occurred during Mg and Al experiments at the ISOLTRAP, lasting Sept. 8th - 16th.

²A UC_x target with a neutron-converter setup, installed on Sept. 28th for IS571 lasting Oct. 3rd - 9th. This target contained no mass markers; previous irradiation of the neutron converter occurred during In experiments at the IDS, lasting Apr. 28th - May 2nd.

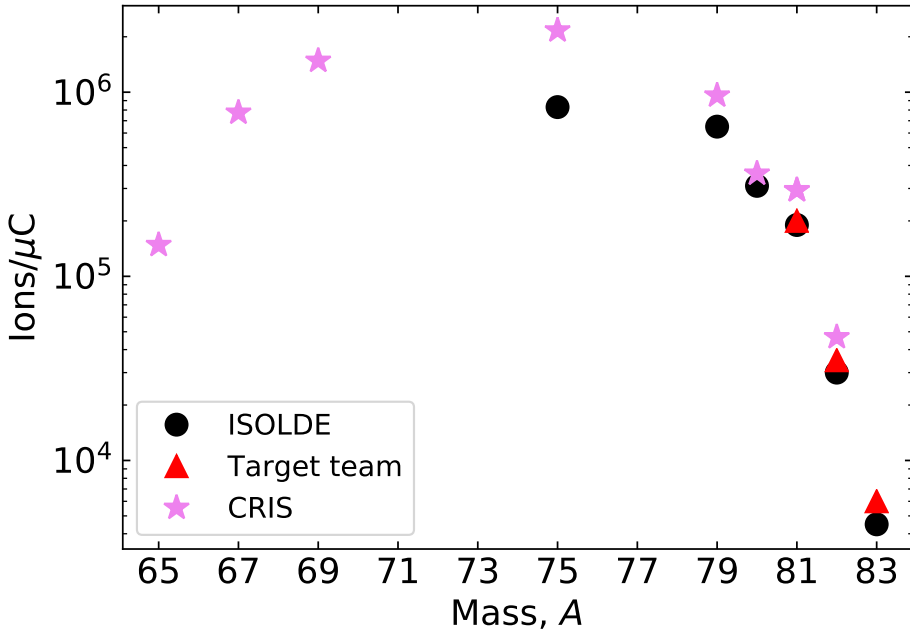


Figure 4.1: Predicted yields for gallium isotopes, from: the ISOLDE yield database [Bal19] (black circles), and the 2015 IS571 experiment as deduced from target-team measurements (red triangles) and the CRIS setup (pink stars).

Efficiency measurements for the 2015 IS571 experiment have been obtained by comparing the number of ions that enter the CRIS beamline to the number that eventually reach a CRIS MCP detector. Due to its status as a mass marker for the UC_x -target setup used in this experiment, ^{71}Ga reference measurements have been utilised for this exercise because any associated isotope beam had been considered to purely consist of this species. ^{71}Ga yields at the CRIS experiment are taken from the most intense resonance featuring in a reference measurement (third right-most peak; see Figure 4.5), and additionally scaled to account for the activity observed at the other resonances (according to the applicable angular-momentum coupling constants as defined in (2.45)). Measuring the yield of an isotope beam just before it enters the CRIS beamline is performed with the use of a FC situated just after the exit of the ISCOOL. Intense stable beams are also normally suppressed before they enter the ISCOOL (and subsequently at the CRIS setup) because otherwise, the MCP detector could experience saturation effects. This is done by applying a pulsing voltage on a beamgate situated before the ISCOOL; a 500 μs temporal length operated at 100 Hz corresponds to a suppression factor of 20 for the isotope beam. With

average FC readings after the ISCOOL of ~ 2 -3 pA and a typical ion-count rate of $\sim 300 - 600 \text{ s}^{-1}$ on the CRIS MCP detector, an average overall experimental efficiency of $\sim 1/1000$ was observed. This is applicable only for the first half of the experiment; this value gradually depreciated towards a reported $\sim 1/5000$ efficiency for the end of the gallium experiment.

Estimated yields of other measured isotopes throughout this experiment are displayed in Figure 4.1; due to the gradual efficiency decline half-way through the experiment, the efficiency used for a particular isotope is taken from the closest ^{71}Ga reference measurement. Within a factor of ~ 3 , a good agreement is obtained between CRIS values with ISOLDE and target-team values. No efficiency measurements are available for the 2017 IS571 experiment because of a lack of FC measurements after the ISCOOL taken throughout.

4.1.2 For francium isotopes

Data relevant for the full laser- and decay-spectroscopy analysis of ^{214}Fr collected during the 2014 IS471 experiment at the CRIS setup are presented in Table 4.2. Akin to the protocol in Chapter 4.1.1, scan numbers correspond to an identifier for the raw-data files; those enclosed in square brackets represent an analysis on a combined data set, with reasons explained in Chapter 7.1.2. Entire data-file names are instead given for the ^{214}Fr decay-spectroscopy measurements³.

Table 4.2: A summary of francium isotope laser- and decay-spectroscopy measurements performed at the CRIS setup, relevant for this thesis. The total time spent collecting data for each isotope is displayed to the right.

2014: IS471			
A	Type	Scan #	Time
214	Laser	[705,706,710], [713,715,716], [726,728], [729,731]	05:39:19
221	Laser	[410,411], [719,720]	00:09:20
			05:48:39
214	Decay	20141129-2307-214Fr-11836.562	00:30:00
		20141129-2346-214Fr-11836.625	00:30:00
			01:00:00

³Additional decay-spectroscopy data for the full $A = 214$ beam can be found in the file with the name: 20141129-2249-214Fr-Def0. Its relevance is explained in Chapter 7.2.3.

4.2 Treatment of raw data from the CRIS setup

The setup of a typical laser spectroscopy experiment at the CRIS experiment is usually designed to have one measurement device for every observable which needs to be tracked with time. This segregation is apparent in the data-file layouts displayed in Figure 4.2, chiefly for: the resonant laser and stabilised HeNe reference laser wavenumber information (wavemeter_ds: $\bar{\nu}_L$ and $\bar{\nu}_D$), the number of observed ion counts (cris_ds: C), and the ISCOOL voltage (iscool_ds: V). Starting with this raw data, the following considerations are important for the transformation of raw data into a format in which hyperfine observables can be reliably extracted:

Timestamp	Resonant Wavenumber, $\bar{\nu}_L$	Reference Diode, $\bar{\nu}_D$
87320996.5879087	11994.319966599	15798.0124695895
87320996.7044039	11994.319966599	15798.0124695895
87320996.82791	11994.319966599	15798.0124695895
87320996.9310403	11994.319966599	15798.0124695895
87320997.0620928	11994.319966599	15798.0124699258
87320997.1185391	11994.3199634229	15798.0124699258
87320997.1560349	11994.3199634229	15798.0124699258
87320997.266125	11994.3199634229	15798.0124699258

Timestamp	Counts, C	
87320996.5878952	0	
87320996.5979073	0	
87320996.5991492	0	
87320996.6091499	0	
87320996.6147869	0	
87320996.6247754	0	
87320996.6347768	0	
87320996.6460354	0	

Timestamp	ISCOOL Voltage, V	
87320997.6545978	39995.4346	
87321002.747262	39995.4237	
87321007.8157821	39995.3451	
87321012.9105959	39995.3915	
87321017.9997027	39995.4223	
87321023.083312	39995.4167	
87321028.1569152	39995.385	
87321033.224144	39995.4167	

Figure 4.2: Typical raw-data formats for wavemeter_ds, cris_ds, and iscool_ds information during a standard CRIS experiment. The above tables display a sample of eight lines for each data file belonging to measurement number #43 during the 2017 IS571 experiment.

- I Since the CRIS measurement devices usually run with different time epochs, the timestamps (located in the first column) for each data file will undoubtedly differ from one another. Thus, one of the first acts of raw data treatment is to make sure that each ion count entry is matched with a corresponding laser wavenumber(s) and ISCOOL voltage at that particular instant in time. One time-synchronisation⁴ method is outlined in Ref. [Gro17a]. The length of time taken to observe C counts for a particular entry should also be determined from their respective timestamps, which will be utilised later on in the data treatment.
- II Vigilance should also be exercised for any wavemeter drifts of the laser system which can seriously affect any isotope-shift interpretations. It explains why a stabilised HeNe reference laser is normally included within a laser setup ensemble and is used to monitor any such deviations by tracking its wavenumber $\bar{\nu}_D$; see Figure 5.3 for those which manifested throughout the 2015 IS571 experiment. By comparing to a known precise transition⁵, the difference is applied as a point-by-point correction to the corresponding $\bar{\nu}_L$ value. Despite this action, other variations might still exist and facilitates the need for either performing regular reference isotope measurements (see Chapter 5.2) or including other additional corrections [Gro17a].
- III Analyses of hyperfine spectra are also usually done with respect to frequency space. The conversion of $\bar{\nu}_L$ (in cm^{-1}) into f_L (in MHz) is given by:

$$f_L = \frac{n c \bar{\nu}_L}{10000}, \quad (4.1)$$

where n represents a frequency multiplication factor related to the method of obtaining the laser light required for the transition⁶ under study, and c represents the speed of light⁷. Additionally, frequency values from (4.1) need to be Doppler shifted to obtain frequencies observed with respect to the accelerated beam f , by using (3.7) and (3.8).

⁴If analysis codes are written in Python, the use of the `np.digitize` and `np.bincount` commands are strongly recommended.

⁵The $5s \rightarrow 3p$ in Ne I is chosen as the reference, with $\bar{\nu} = 15798.0031 \text{ cm}^{-1}$ [NIS18].

⁶Common values include: 1 for fundamental, 2 for frequency-doubling, 3 for frequency-tripling, and 4 for frequency-quadrupling.

⁷Considering that hyperfine interactions represent energy perturbations at least 10^6 smaller than the atomic fine structure, using the (commonly-utilised ‘undergraduate’) value of $c = 3 \times 10^8 \text{ ms}^{-1}$ would be insufficient! The value $c = 299792458 \text{ ms}^{-1}$ represents the minimum precision that should be used; this can be defined in Python using `scipy.constants`.

IV The final procedure for the raw data should be to bin it with a sufficient frequency width to resolve the observed hyperfine structures. Especially for isotope measurements involving low yields, it may be necessary to sum several data sets together in order to distinguish these resonances from any occurring background rate. Working with ion-count rates R is more convenient because the total amount of time T spent at a particular frequency bin may not be constant throughout:

$$R = \frac{C}{T}, \quad \text{and} \quad \sigma_R = \frac{\sigma_C}{T}, \quad (4.2)$$

where the error on the count rate $\sigma_C = \sqrt{C}$ is borne from the underlying Poisson nature of counting experiments [Kno10]. The cases displayed in (4.2) highlight the absurdity of potential uncertainties which would otherwise be consistent with negative count rates. In order to avoid problems with the cost function calculation (further detailed in Chapter 4.3), $\sigma_C = \sqrt{C+1}$ is usually used instead and addresses the cases where measuring a particular frequency point is associated with zero counts.

4.3 Choice of cost function

The extraction of information from experimental data requires a reasonable model $f(x_i)$ which considers all occurring atomic effects described in Chapter 3.2.1. Although lineshape choices will be discussed next in Chapter 4.4, defining a measure of how well it fits experimental data is first given and is commonly referred to as the *cost function*. The most recognisable is the reduced chi-squared per degree of freedom χ_ν^2 , defined as:

$$\chi_\nu^2 = \frac{1}{\nu} \sum_{i=1}^N \left(\frac{y_i - f(x_i)}{\sigma_{y_i}} \right)^2, \quad (4.3)$$

which for the case of usual laser-spectroscopy measurements, is calculated from a spectrum containing N data points each corresponding to a count rate $y_i \pm \sigma_{y_i}$ sampled for a given frequency step x_i . The degree of freedom $\nu = N - m$ is equivalent to the difference between N with the number of fitted parameters contained within the chosen function m . Alternative descriptions such as the log-likelihood can also be used, however simulations in Ref. [Gin18] show that there is a negligible difference in the overall result if used in high-statistics

data sets. With most measurements⁸ presented in this thesis considered to fall under the high-statistics regime, all measurement fits have been performed with a reduced chi-squared analysis as defined in (4.3). It is remarked that while the ^{214}gFr measurements presented in Chapter 7.1.2 could be considered as low-statistics datasets, the large minimum uncertainties imposed by the lack of linewidth resolution completely negates any improvements that might be gained by using a different methodology to (4.3).

Non-standard minimisation techniques, such as those which use Bayesian statistics, are normally used if additional uncertainties need to be taken into consideration. This has been used for performing a modified King plot-analysis with francium isotope-shift values, in order to take into account the large correlation between the extracted gradient and intercept for a linear relation. A further mention of this is provided in Chapter 7.4.

4.4 Lineshape profiles for laser spectroscopy

Furthermore to the procedures outlined in Chapter 4.2, it is important to fit observed hyperfine resonances with a model which considers all the occurring atomic effects from Chapter 3.2.1. The main lineshapes used for the data analysis in this thesis all derive from commonly recognised spectral profiles: Gaussian, and Lorentzian. Serving partly as a reminder to the aforementioned chapter, Gaussian G profiles are used to model the Doppler broadening with a distribution at any given position x for a resonance observed at position x_0 defined by:

$$G(x, x_0, \sigma) = \frac{1}{\sqrt{2\pi}\sigma} e^{-(x-x_0)^2/2\sigma^2}, \quad (4.4)$$

where its Full Width at Half Maximum (FWHM) is defined by $f_G = 2\sigma\sqrt{2\ln 2}$. Natural linewidths and power broadening effects on the other hand, are Lorentzian L in nature. Using the same spatial considerations as defined above, this profile is expressed as:

$$L(x, x_0, \gamma) = \frac{\gamma}{\pi} \frac{1}{(x - x_0)^2 + \gamma^2}, \quad (4.5)$$

⁸The least intense measurement from the gallium data set is of ^{82}Ga , with a peak count rate of $\sim 7 \text{ s}^{-1}$; equivalent to ~ 500 raw counts.

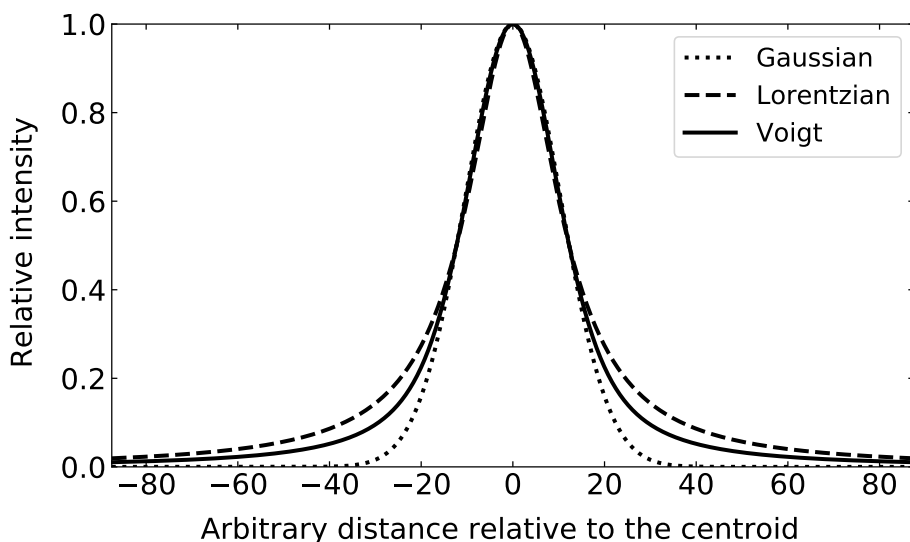


Figure 4.3: Normalised Gaussian (dotted line), Lorentzian (dashed line) and Voigt¹⁰ (full line) profiles. Each displayed lineshape has a FWHM of 25 MHz.

with a FWHM of $f_L = 2\gamma$. For low-resolution laser-spectroscopy measurements, one common approximation that can be supposed is that their $\sim$ GHz linewidths predominantly arise from Doppler broadening; thus to an approximation, any observable hyperfine resonances could be fitted with pure Gaussian profiles. Low-resolution measurements of ^{221}Fr and ^{214}Fr taken during the 2014 IS471 experiment (see Chapters 7.1.1 and 7.1.2) have utilised this lineshape assumption, due to the broadband nature of 417.2-nm light provided by the RILIS group at the ISOLDE facility.

4.4.1 Voigt lineshapes

One of the most commonly utilised lineshapes in laser spectroscopy involves the convolution of a Gaussian and a Lorentzian profile as defined in (4.4) and (4.5) respectively, resulting in the Voigt profile. While this represents a lineshape which can be used to model all the atomic manifestations as mentioned above, its evaluation is normally an incredibly complex task. Conveniently however, it can be related to the real part of the following Faddeeva function W :

¹⁰The individual Gaussian and Lorentzian contributions both have equivalent FWHM's of ~ 15 MHz, yielding a total FWHM of 25 MHz for the displayed Voigt profile.

$$V(x, x_0, \sigma, \gamma) = \frac{1}{\sqrt{2\pi}\sigma} \Re \left[W \left(\frac{(x - x_0) + i\gamma}{\sqrt{2}\sigma} \right) \right], \quad (4.6)$$

in which x , x_0 , γ , and σ have already been defined in Chapter 4.4. Its FWHM likewise is also difficult to evaluate, however one interpretation defines it as:

$$f_V \approx 0.5346 f_L + \sqrt{0.2166 f_L^2 + f_G^2}, \quad (4.7)$$

which is accurate up to 0.02% with respect to the individual Gaussian and Lorentzian contributions [Oli77]. Figure 4.3 shows the geometrical differences exhibited between (4.4), (4.5), and (4.6) each with similar total FWHM's. High-resolution measurements of ^{221}Fr taken during the 2014 IS571 experiment (see Chapter 7.1.1), for which the CRIS setup was able to provide 417.2-nm light, have been fitted with these Voigt profiles.

4.4.2 Crystal Ball lineshapes

As already alluded to in Chapter 3.3.2, the 2015 IS571 gallium experiment on encountered occurrences of high-frequency lineshape distortions associated with every observable resonance. This effect is thought to originate from Alternating-Current (AC) Stark-shift effects, also known as the Autler-Townes effect [Aut55], in which an oscillating electric field can change the lineshape by interacting with the transition frequency. For this laser-spectroscopy scenario, it is the electric field generated by the high-power 1064-nm light that can distort the positions of the lower energy levels for the excitation steps. This has previously featured in CRIS-setup measurements of francium and copper isotopes, which could be remedied by sufficiently delaying the laser pulses required for ionisation, with respect to the laser light probing the transition step [Gro17d]. For the gallium-transition scheme used however, a $t_{1/2} = 5.7(6)$ ns lifetime of the intermediate $4s^2 5s \ ^2S_{1/2}$ state [Saf06] (see Figure 3.5) does not allow for sufficient laser-delay manipulation. With the aim of trying to measure the most neutron-rich species [Coc13a], any laser-power reductions in order to alleviate these effects were not seen as viable options. Therefore, attention turned to finding a suitable model in which these effects could be appropriately modelled and allow for the accurate extraction of the hyperfine observables.

One lineshape considered involved convoluting Gaussian distributions from (4.4) with a power-law tail, colloquially referred to as the *Crystal Ball*¹¹ function C . With x , x_0 , and σ already defined in (4.4), its full definition is defined as:

$$C(x, x_0, \sigma, \alpha, n) = N \cdot \begin{cases} \exp\left(-\frac{(x-x_0)^2}{2\sigma^2}\right), & \text{for } \frac{x-x_0}{\sigma} > -\alpha \\ \left[\left(\frac{n}{|\alpha|}\right)^2 \cdot \exp\left(-\frac{|\alpha|^2}{2}\right)\right] \left[\frac{n}{|\alpha|} - |\alpha| - \frac{x-x_0}{\sigma}\right]^{-n}, & \text{for } \frac{x-x_0}{\sigma} \leq -\alpha \end{cases} \quad (4.8)$$

, in which both C and its first derivative are both continuous. N represents some normalisation factor and the other two as-of-yet undefined parameters, α and n , encapsulate the asymmetric nature of this function: α ¹² to symbolise the boundary which joins these two distributions together, and n to define the order of the power-tail. Graphical representations of how these two parameters

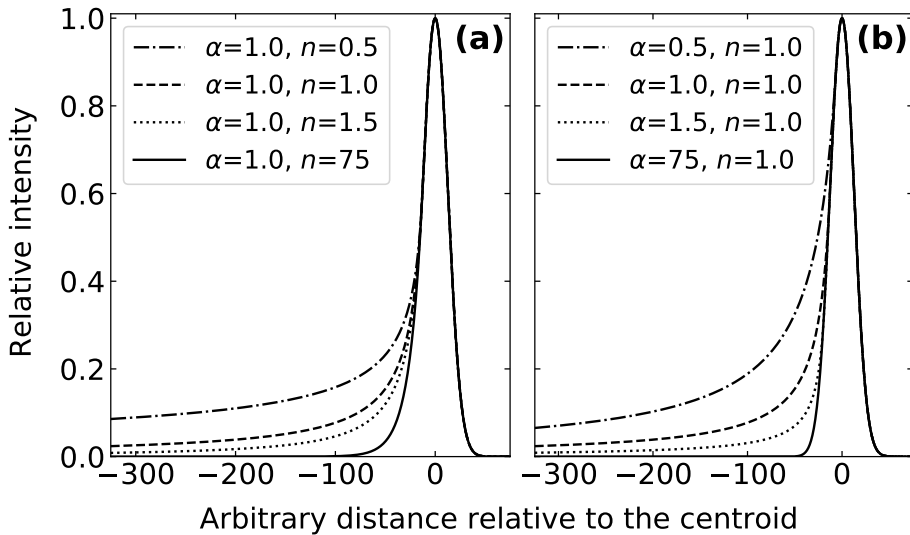


Figure 4.4: The changes in the shape of a normalised Crystal Ball profile by varying the: (a) α , and (b) n asymmetry parameters. Each displayed lineshape has a FWHM of 30 MHz.

¹¹This originates from an alike collaboration at the Stanford Linear Accelerator Centre. See Refs. [Ore80] (Appx. D), [Gai82] (p. 178), and [Skw86] (Appx. E) for more details.

¹²By default, a Crystal Ball function defined by (4.8) features a power-law tail on the left-hand side. To obtain one with a right-handed asymmetry, a sign change needs to be implemented for all cases of x and x_0 in (4.8), both in the equation and condition descriptions.

affect the overall lineshape can be seen in Figure 4.4. The Crystal Ball function had already been used by the CRIS collaboration for characterising the energy loss of α particles travelling through carbon foils at the DSS2.0 setup [Lyn17]. Alpha-decay spectroscopy studies performed at the ISOLDE facility have also similarly utilised these lineshapes in their analyses [Cub18; Cub19]. Even though AC Stark-shift effects represent something wholly different, the Crystal Ball function was seen as an appropriate empirical choice at the time of analysing the 2015 IS571 data set.

Figure 4.5 provides fit comparisons between using Crystal Ball and Voigt lineshapes for a variety of ^{71}Ga reference measurements collected throughout the 2015 IS571 experiment. A factor of ~ 5 reduction in the obtained χ^2_ν values is generally obtained if the Crystal Ball function is utilised instead of usual Voigt profiles. Subsequent extracted hyperfine parameters displayed in Figure 4.6 additionally reveals not only consistency with respect to existing tabulated values for ^{71}Ga in Ref. [Che10b], but also an invariance between the two lineshape profiles to within statistical uncertainties. Based on this new-found confidence for its usability, Crystal Ball lineshapes are thus used for fitting all hyperfine measurements of gallium isotopes collected during the 2015 IS571 experiment. A further characterisation with the extracted hyperfine parameters for the entire ^{71}Ga data set using Crystal Ball lineshapes for fitting is given in Chapter 5.2. While a small variation can be seen with respect to the extracted centroid-frequency differences between the two lineshape profiles, this is not significant to within statistical uncertainties. An average ~ 6 MHz difference is observed, however this does not impact on any isotope-shift measurements because this difference would undoubtedly cancel out.

Additional studies had also been conducted to see if reducing the intensity of any of the laser-light pulses involved in the gallium-transition scheme (see Figure 3.5) would effectively remove the presence of these lineshape distortions. Although using less-intense powers for the 1064-nm light may seem the most logical answer, the graphical analysis presented in Figure 4.7 shows that this did not produce any significant results; a lineshape-distortion suppression of 38(17)% could only be achieved by reducing the pulse energy from 40 mJ/pulse to 23 mJ/pulse. A constant FWHM of ~ 100 MHz was also reported for the three different powers tested. Investigating powers with < 23 mJ/pulse could not be made without adversely affecting the ionisation rate from within the UHV region of the CRIS setup. A better removal of up to 72(27)% could be achieved by instead reducing the power intensity of 417.2-nm light from 170 mW to 30 mW; this is graphically displayed in Figure 4.8. Additionally in contrast to the observation in Figure 4.7, the observed FWHM is reduced by 10% to ~ 90 MHz. Despite these relatively modest advantages, the observed Signal-to-Background (StB) ratio was detrimentally affected by up to a factor

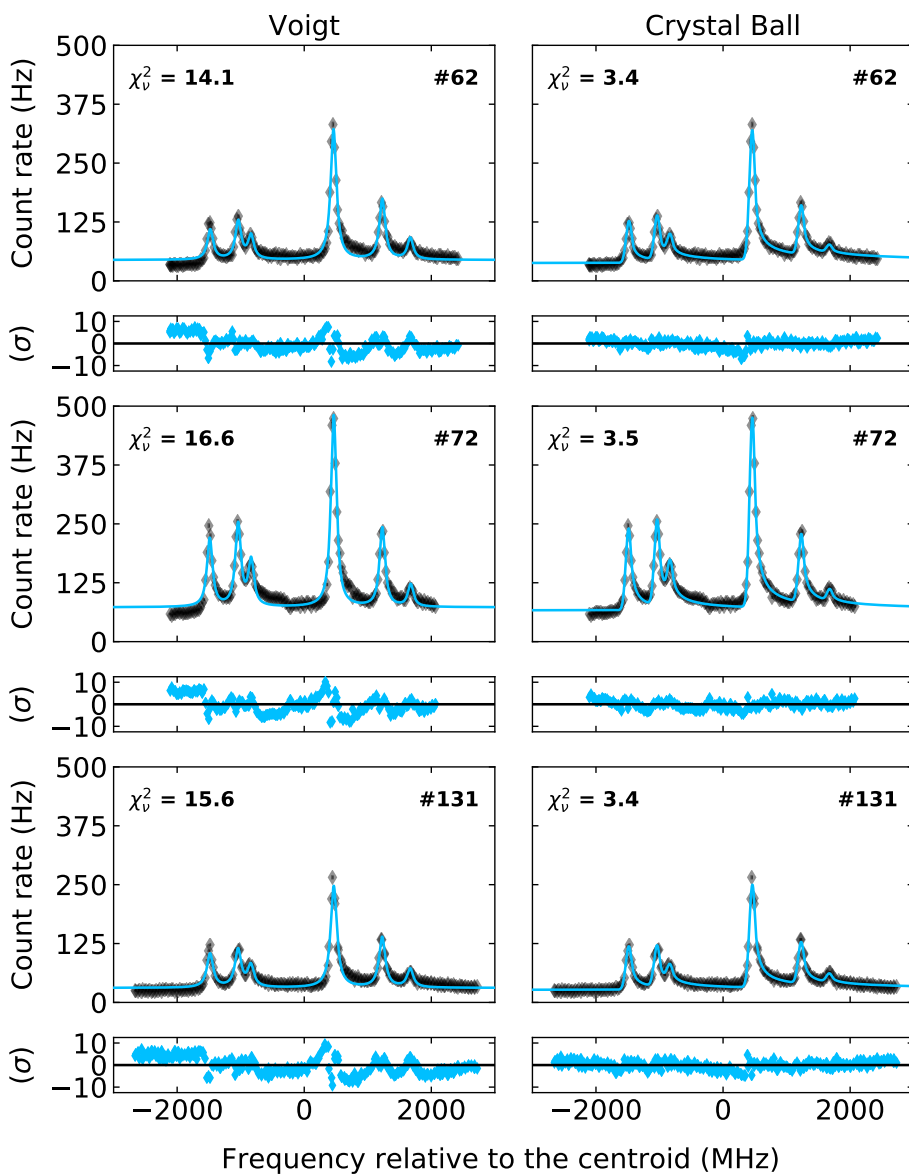


Figure 4.5: A comparison of fitting with: Voigt (left column), and Crystal Ball (right column) profiles, for selected 2015 IS571 ^{71}Ga measurements (#62, #72, and #131). Residuals representing the normalised σ (error-bar) difference between a fit with the data points are given below each spectrum; χ^2_ν values are also displayed.

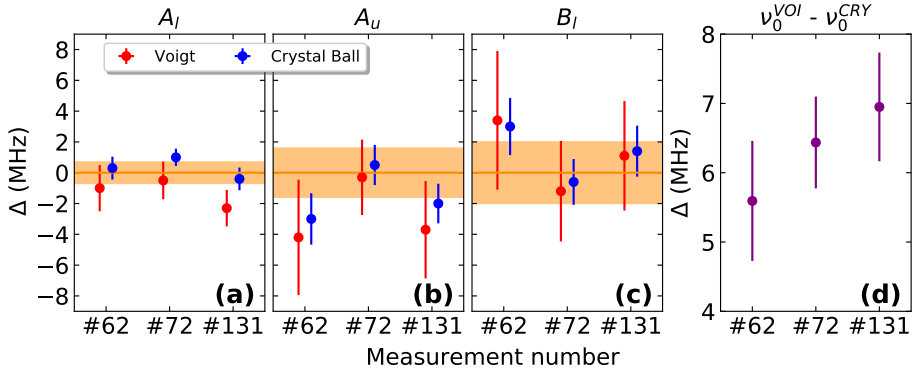


Figure 4.6: A comparison of the differences between extracted: (a) A_I -, (b) A_u -, and (c) B_I -hyperfine observables from fitting the hyperfine spectra in Figure 4.5 with Voigt (red) and Crystal Ball (blue) profiles, with the tabulated values from Ref. [Che10b] (orange lines). Centroid-frequency differences between extracted values from the Voigt and Crystal Ball profiles (purple) are given in (d).

of ~ 3 . Considering the main aim of measuring the most exotic neutron-rich gallium isotopes for the 2015 IS571 experiment [Coc13a], these power reductions were not enforced in order to provide a better chance in measuring these species.

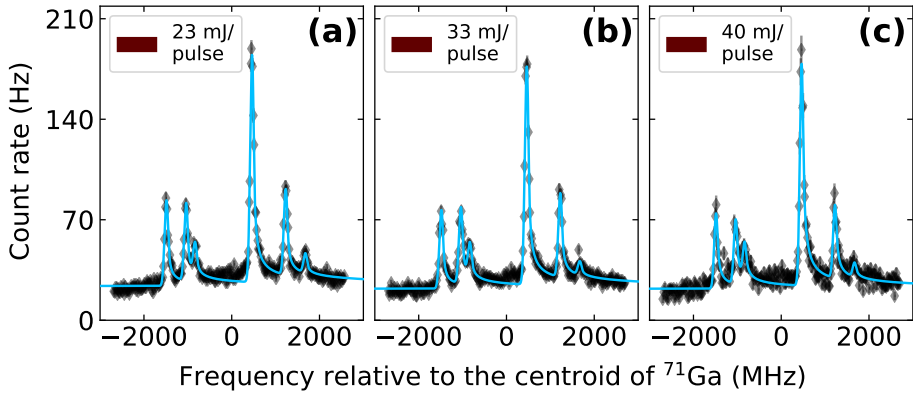


Figure 4.7: Selected ^{71}Ga measurements obtained using: (a) 23 mJ, (b) 33 mJ, and (c) 40 mJ 1064-nm pulse energies, from the Litron laser setup during the 2015 IS571 experiment. 30 mW and 100 μJ / pulse of 417.2-nm and 639-nm light respectively, have been used for each scan. Crystal Ball lineshapes have been utilised for fitting the spectra.

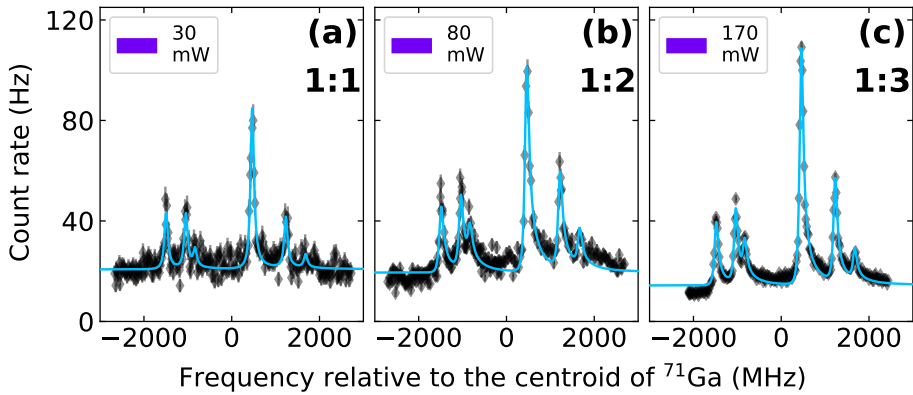


Figure 4.8: Selected ^{71}Ga measurements obtained using: (a) 30 mW, (b) 80 mW, and (c) 170 mW 417.2-nm powers, from the M-Squared SolsTiS setup during the 2015 IS571 experiment. $100\ \mu\text{J}$ / pulse and 35 mJ / pulse of 639-nm and 1064-nm light respectively, have been used for each scan. Crystal Ball lineshapes have been utilised for fitting the spectra. Scale factors apply to the count rate for a particular spectra.

4.4.3 An asymmetry parameter for lineshapes

Following the analysis and publication of the data set from the 2015 IS571 experiment in **Article I** (see Chapter 5.1), another asymmetric description had been encountered for the modelling of lineshape distortions observed at the CRIS setup. As described in Ref. [Sta08], (4.4), (4.5), and (4.6) can be modified by substituting the Gaussian or Lorentzian width parameters, for the purpose of this explanation denoted by γ_0 , with γ_ν :

$$\gamma(\nu) = \frac{2\gamma_0}{1 + \exp\{a(x - x_0)\}}, \quad (4.9)$$

where a is defined as the asymmetry parameter, skewing towards higher frequencies for negative values. When $a = 0$, (4.9) simply reverts back to the original substituted width parameter. Coincidentally it has already been utilised in the aforementioned reference for fitting infrared absorption spectra, which is considered from within the family of spectra-related phenomena like that observed in hyperfine-resonance spectroscopy (see Chapters 4.4 and 4.4.1). Akin to Figure 4.5, Figure 4.10 compares this new asymmetric definition with respect to the performance of Voigt and Crystal Ball profiles previously utilised

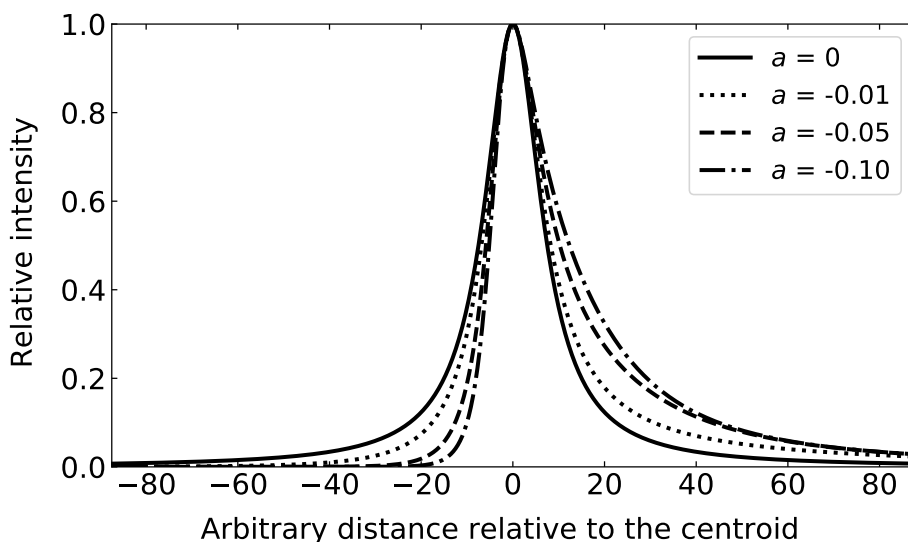


Figure 4.9: The effect of the asymmetry parameter a on the lineshape of a Lorentzian distribution. All lineshapes displayed in the figure have a FWHM of 15 MHz; the reasoning for choosing a Lorentzian lineshape for this display is explained further in the text.

beforehand, for ^{71}Ga reference measurements taken during the 2017 IS571 experiment. Only asymmetric-Lorentzian profiles are considered because a cursory investigation involving fitting with asymmetric Voigt profiles revealed negligible ≤ 1 MHz Gaussian contributions for all measurements.

While this additional modification offers a factor of ~ 2 reduction in the χ^2_ν values for available data compared with Crystal Ball profiles, it is remarked that these values are unusually high with respect to those in Figure 4.10. One explanation had been due to inherent power-broadening effects as a result of using an injection-seeded laser system as described in Chapter 3.3.2; Figure 4.12 shows that dealing with $\sim \text{mW}$ powers with this new laser setup resulted in massive power-broadening effects of the $\sim \text{GHz}$ order. The observation of clearly recognisable hyperfine structure akin to that in Figure 4.5 could actually be obtained by using $\sim \mu\text{W}$ power densities, an action which was not fully realised at the beginning of the 2017 IS571 experiment. The power values recorded for each spectrum in Figure 4.12 however, only represent approximate values. This is because of problems encountered during the start of the 2017 IS571 experiment which involved: yield-measurement attempts with a UC_x target containing no mass marker, initial confusion with the dominant power-

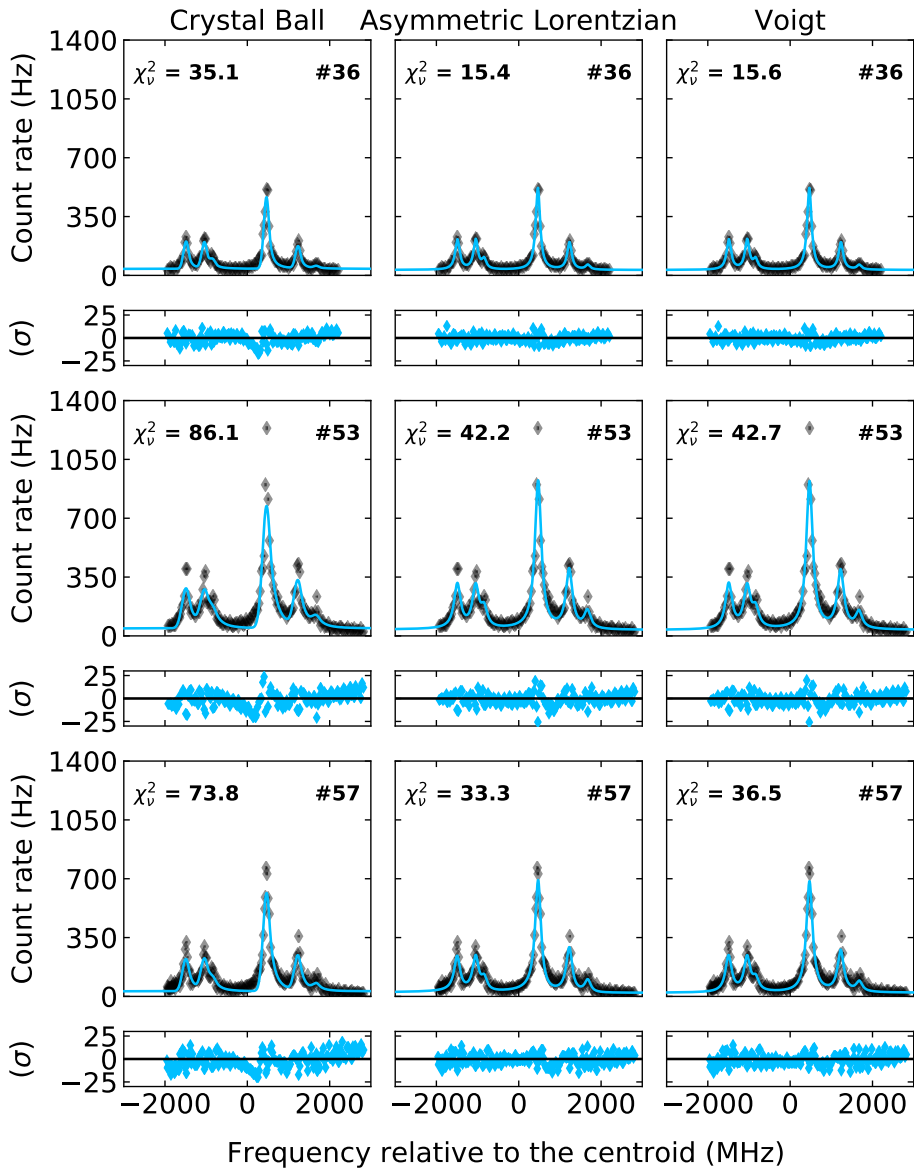


Figure 4.10: A comparison of fitting with: Crystal Ball (left column), asymmetric-Lorentzian (centre column), and Voigt (right column) profiles, for selected 2017 IS571 ^{71}Ga measurements (#36, #53, and #57). Residuals representing the normalised σ (error-bar) difference between a fit with the data points are given below each spectrum; χ^2_ν values are also displayed.

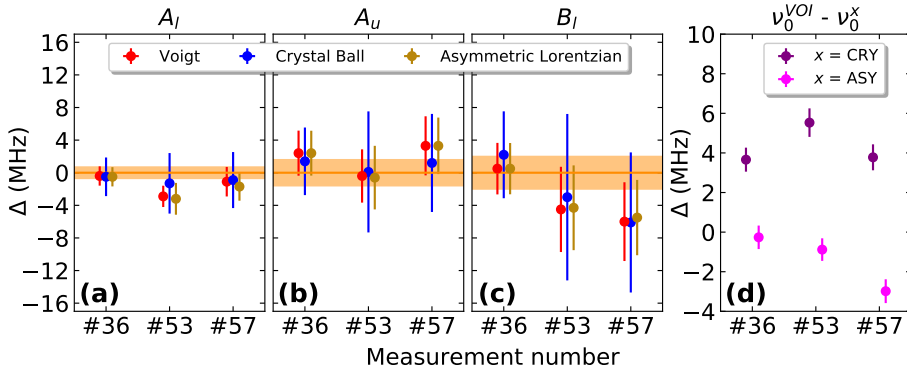


Figure 4.11: A comparison of the differences between extracted: (a) A_I -, (b) A_u -, and (c) B_I -hyperfine observables from fitting the hyperfine spectra in Figure 4.10 with Voigt (red), Crystal Ball (blue), and asymmetric-Lorentzian (yellow) profiles, with the tabulated values from Ref. [Che10b] (orange lines). Centroid-frequency differences between extracted values from the Voigt profile with the other used profiles (purple and pink) are given in (d).

broadening effects mentioned beforehand, and attempting isotope measurements with a progressively failing target. As such, information for some isotope measurements taken during the 2017 IS571 experiment is sparsely recorded in the experimental logbook, which made it difficult to be able to comprehensively study these power-broadening effects in more detail. Any future gallium experiment should make the detailed study of how gallium hyperfine structures are affected with different laser-power densities, an important priority. One of the most striking effects between Figures 4.8 and 4.12 is the obtainable StB ratios from using the two methods to produce pulsed 417.2-nm light as described in Chapter 3.1.2. While a maximum ratio of 6(1) had been achieved using a laser power of 170 mW during the 2015 IS571 experiment (see Figure 3.6(c)), ratios of up to 30(2) could be achieved in the 2017 IS571 experiment with pulsed energies almost a factor of 10^4 less intense than beforehand.

Overall χ^2_ν values associated with asymmetric-Lorentzian profiles are however, similar to those obtained using Voigt profiles, but with a slight preference for the former lineshape. This is due to a much reduced AC Stark-shift effect; it had been discovered (and not considered during the 2015 IS571 experiment) that only delaying the 1064-nm light, instead of along with the 639-nm light (see Chapter 4.4.2), by at least 70 ns majorly removed the presence of these spectral effects. A comprehensive study however, could not be made because of laser-power densities and delays not being recorded for some of the measurements involved. With respect to the extracted hyperfine parameters for each lineshape in Figure

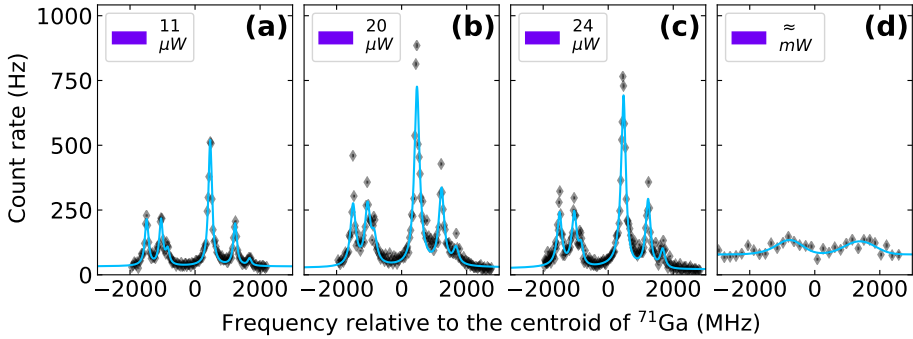


Figure 4.12: Selected ^{71}Ga measurements obtained using: (a) $11\ \mu\text{W}$, (b) $20\ \mu\text{W}$, (c) $24\ \mu\text{W}$, and (d) mW-order 417.2-nm powers, from the injection-seeded Ti:Sa laser setup during the 2017 IS571 experiment. 639-nm and 1064-nm pulse energies and powers are unknown for these scans, however they are assumed to be the same values used in the 2015 IS571 experiment because there are no documented value changes in the CRIS-setup logbook. Asymmetric-Lorentzian lineshapes have been utilised for fitting the spectra.

4.11, they are consistent not only with previous literature values [Che10b] but also with each other, analogous to the conclusions of the previous 2015 IS571 study in Figure 4.6. While the centroid-frequency differences between Voigt and Crystal Ball profiles are seen to be more-or-less constant, there is a slight trend when compared with asymmetric-Lorentzian profiles. Further investigations were not done however, because of the lack of isotope measurements made in the 2017 IS571 experiment, as evidenced in Table 4.1. Nonetheless, asymmetric-Lorentzian profiles have been used to fit hyperfine-structure measurements taken throughout the 2017 IS571 experiment, in order to make sure that any potential asymmetry in these isotope measurements have been taken into account.

4.5 Combining multiple measurements of an observable

Unless a particular isotope n requires a substantial length of time for its successful measurement, because it is either produced with a low yield or initially indistinguishable from high background levels, it is not necessary to sum all the data. Instead for a set of N hyperfine-observable measurements each with uncertainties, $(\lambda_n \pm \sigma_n)$, it is usually customary to represent the final reported value $\bar{\lambda}_n$ with a weighted mean:

$$\bar{\lambda}_n = \frac{\sum_1^N \lambda_n w_n}{\sum_1^N w_n} = \frac{\sum_1^N \lambda_n / \sigma_n^2}{\sum_1^N 1 / \sigma_n^2}, \quad (4.10)$$

with $w_n = 1/\sigma_n^2$ as the usual choice of weight. with a standard error of the weighted mean $\sigma_{\bar{\lambda}_n}$ of:

$$\sigma_{\bar{\lambda}_n} = \sqrt{\frac{1}{\sum_i^N w_n}} = \sqrt{\frac{1}{\sum_i^N 1/\sigma_n^2}}. \quad (4.11)$$

However in order to correct for dispersion effects, the error in (4.11) is scaled with respect to the total reduced chi-squared χ_ν , to result in a standard error of the weighted mean $\hat{\sigma}_{\bar{\lambda}_n}$:

$$\hat{\sigma}_{\bar{\lambda}_n} = \sigma_{\bar{\lambda}_n} \sqrt{\chi_\nu} = \sqrt{\frac{\sum_i^N (\lambda_n - \bar{\lambda}_n) / \sigma_n^2}{(N-1) \sum_i^N 1/\sigma_n^2}}, \quad (4.12)$$

which is what is used to report on the final statistical uncertainty for the measurement of a particular observable.

4.6 Summary

The final set of tools required for the thorough interpretation of hyperfine spectra measured at the CRIS setup has been given. A repertoire of lineshapes has also been described, each of which modelling different nuances of atomic effects that can potentially manifest whenever a hyperfine-structure measurement is being taken. Alongside theoretical nuclear physics concepts in Chapter 2 and knowledge of the CRIS technique in Chapter 3, probing the magical characters of doubly closed-core nuclei can now commence.

Chapter 5

Nuclear magicity near ^{56}Ni

Possessing both $N = Z = 28$, ^{56}Ni is the first doubly-magic nucleus entirely borne from the coupling between the spin-orbit interaction and the nuclear potential. The characterisation of fundamental ground-state-property trends

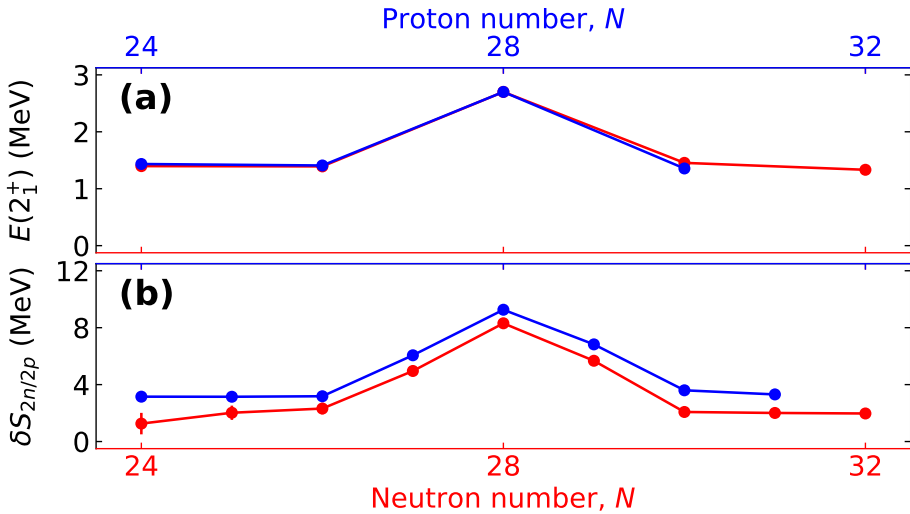


Figure 5.1: (a) Energies for the first excited 2^+ state. (b) Two-nucleon-separation energy differences between $N, N+2$ and $Z, Z+2$, for the neutrons and protons respectively. In effect, trends for $Z = 28$ (Ni) isotopes (red) and $N = 28$ isotones (blue) feature in both plots. Data are taken from Ref. [NND18].

in its vicinity therefore, is vital for confirming the importance of additional corrections to the harmonic oscillator, as discussed in Chapter 2.1.1 (see (2.4) and Figure 2.1). Within the discussion of this same section, a glance at the isotopic and isotonic trends for $E(2_1^+)$ and $S_{2n/2p}$ energies in Figure 5.1 reveals distinct peaks at $N, Z = 28$. With a magical signature already established, this provides the groundwork for further investigation into the ground-state properties in order to discern its exact magical nature.

Extensive theoretical studies have already predicted a rather soft ^{56}Ni core [Hon02a; Hon04], which is the basis for the GXPF1 family of shell-model interactions. Experimentally obtained magnetic-dipole moments for all neighbouring one-particle/one-hole systems: ^{57}Cu [Coc09], ^{57}Ni [Oht96], ^{55}Co [Cal73], and ^{55}Ni [Ber09], can be favourably replicated using this interaction. Recent electromagnetic moment comparisons for $^{53}_{26}\text{Fe}_{27}$ also confirm its soft nature [Mil17]. While these measurements are important to structurally characterise ^{56}Ni , there is an equal, if not greater, need for electric-quadrupole moments. This is due to their sensitivity to any collective effects which may manifest in regions near suspect shell closures, such as ^{78}Ni [Gro17b] (see Chapter 6). Trends in the odd- A $_{29}\text{Cu}$ [Vin11b] and $_{31}\text{Ga}$ [Che10b; Pro12] isotope between $N = 28$ to $N = 40$ indicate increased collectivity effects towards the former neutron number, *id est* near ^{56}Ni . However a scarcity of measurements near its immediate vicinity, especially for the $_{31}\text{Ga}$ isotopes, prevents any firm statements. This provides the motivation for continuing the investigation of isotopes in this region of the nuclear chart for a complete characterisation.

The essence of **Article I** investigates the ground-state properties of $_{31}\text{Ga}$ isotopes, with a particular attention to isotopes nearing the ^{56}Ni region. Alongside supplementary analyses provided in the successive section, updated insights regarding the magical effects towards these isotopes are provided.

5.1 Article I: Probing the $_{31}\text{Ga}$ ground-state properties in the region near $Z = 28$ with high-resolution laser spectroscopy

Probing the ^{31}Ga ground-state properties near the $Z = 28$ region with high-resolution laser spectroscopy

G. J. Farooq-Smith,^{1,2,*} A. R. Vernon,² J. Billowes,² C. L. Binnersley,²
M. L. Bissell,² T. E. Cocolios,^{1,2} T. Day Goodacre,^{2,3,†} R. P. de Groote,¹
K. T. Flanagan,² S. Franchoo,⁴ R. F. Garcia Ruiz,^{1,2} W. Gins,¹ K. M. Lynch,⁵
B. A. Marsh,³ G. Neyens,¹ S. Rothe,^{2,3} H. H. Stroke,⁶ S. G. Wilkins,² and X. F. Yang¹

¹*KU Leuven, Instituut voor Kern- en Stralingsfysica, Celestijnenlaan 200D,
3001 Leuven, Belgium*

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

³*EN Department, CERN, CH-1211 Geneva 23, Switzerland*

⁴*Institut de Physique Nucléaire d'Orsay, F-91406 Orsay, France*

⁵*ISOLDE, EP Department, CERN, CH-1211 Geneva 23, Switzerland*

⁶*Department of Physics, New York University, New York, New York 10003, USA*

Abstract

Magnetic-dipole and electric-quadrupole moments for $^{65,67,69,75,79-82}\text{Ga}$ are reported using the Collinear Resonance Ionization Spectroscopy (CRIS) technique at the ISOLDE facility, CERN. The moments of ^{65}Ga have been measured for the first time: $\mu = +1.775(3)\mu_N$ and $Q_s = +21.0(15)\text{efm}^2$. These results are compared to shell-model calculations using the GXPF1 and JUN45 interactions and the trends of the moments approaching the region of the doubly magic ^{56}Ni are discussed. Additionally, new values for the change in mean-square charge radii for ^{65}Ga and ^{67}Ga allow investigation into the odd-even staggering in the region below $N = 40$.

* gregoryjames.farooqsmith@kuleuven.be

† Present address: TRIUMF, Vancouver, British Columbia V6T 2A3, Canada

I. INTRODUCTION

Over the past few decades, the investigation of the ground-state properties of isotopes residing in the vicinity of the shell closure $Z = 28$ has attracted significant interest. In the region near the doubly magic isotope $^{78}\text{Ni}_{50}$, there are signatures of shape coexistence: low-energy intruder states in the nearby $N = 49$ isotones [1, 2] and the observation of an intruder 0_2^+ state in ^{80}Ge [3]. A kink at $N = 50$ in the relative mean-square charge radii for the gallium isotopes adds further confirmation to its status as a magic number [4, 5] and several theoretical studies have been dedicated to the study of this magic number [6–9].

Additionally, the reversal of the $\pi 1f_{5/2}$ and $\pi 2p_{3/2}$ orbitals can be attributed to an attractive monopole interaction which strengthens as the neutron $\nu g_{9/2}$ orbital gets filled [10]. This eventually manifests itself with a ground-state spin change and has been observed for the copper and gallium isotope chains: in ^{75}Cu [11] and ^{79}Ga [12] respectively. The monopole tensor interaction has also been important for explaining the triple shape coexistence exhibited in ^{70}Ni [13], along with the presence of proton and neutron excitations across the $Z = 28$ shell gap. The inclusion of these cross-shell excitations in shell-model calculations has recently helped explain the systematics of the nuclear electromagnetic moments for this region, such as the electric quadrupole moments in the manganese isotopes [14]. However, the amount of excitations required in shell model calculations to explain evolutions in shell gaps and single-particle energies still remains to be discovered.

For the neutron-deficient region above $Z = 28$, a sudden increase in the magnetic moment was observed for ^{57}Cu , towards the effective single-particle $\mu(\pi p_{3/2})$ value [15]. While $N, Z = 28$ are still good magic numbers in this region of the nuclear chart, a softer $^{56}\text{Ni}_{28}$ was required to obtain good agreement between experiment and theoretical predictions [16]. However, the electric-quadrupole moments are a better probe of the softness of the core, due to the strong correlation of collectivity with pair scattering across the orbitals. Later studies of the copper quadrupole moments revealed an increasing collectivity as $N = 28$ is approached and reinforced the suggestion of a soft core [17], although they could not reach $N < 29$. Similar studies into these moments for the neutron-deficient gallium isotopes will provide additional insight into this phenomenon.

In this article, we report on the measurements of the radioactive gallium isotopes $^{65,67,69,75,79-82}\text{Ga}$. Newly presented magnetic dipole and electric quadrupole moments for

^{65}Ga are compared to shell-model calculations from recent interactions, helping to characterize the systematics in the neutron-deficient region below $N = 40$. Changes in the mean-square charge radii for $^{65,67}\text{Ga}$ are also presented alongside literature values for other gallium isotopes. Where available, data are compared to literature values, showing good agreement and a similar precision for the moments and changes in the mean-square charge-radii.

II. EXPERIMENTAL METHOD

Beams of radioactive gallium isotopes were produced at the ISOLDE facility in CERN [18] by first impinging 1.4 GeV protons from the Proton-Synchrotron Booster [19] onto a solid tungsten neutron converter [20]. Neutrons produced from the resulting spallation processes irradiated a thick UC_x target to induce fission events. From this, gallium isotopes diffused

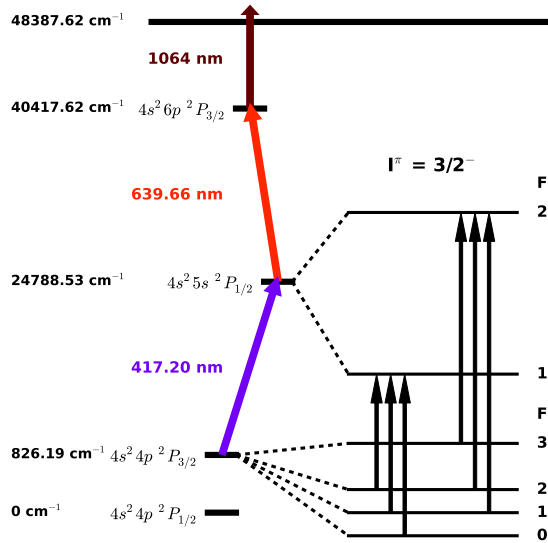


FIG. 1. (Color online) A diagram of the resonance ionization scheme used for neutral gallium, including the allowed hyperfine transitions (not to scale) for the 417.2-nm transition, assuming $I^\pi = 3/2^-$.

out of the target material and effused towards a high temperature (~ 2200 K) tantalum ion source, where they were laser-ionized using the Resonance Ionization Laser Ion Source (RILIS) [21], and extracted at 30 keV. The mass regions of interest were selected using the high-resolution mass separator [18] and subsequently cooled and bunched using the ISOLDE cooler buncher (ISCOOL) [22, 23].

After re-accelerating to 30 keV, the ion bunches were electrostatically deflected into the collinear resonance ionization spectroscopy (CRIS) beamline [24–26] and passed through a Charge Exchange Cell (CEC). This was filled with a potassium vapor (held at 460–470 K), which neutralized the ion bunches with a neutralization efficiency of 2.5%. At this temperature, charge-exchange calculations based on the work of Rapp and Francis [27] showed that the ground and metastable states (as shown in Fig. 1) are roughly equally populated. Any non-neutralized components remaining were deflected away. After passing through a differential-pumping region (just after the CEC), the neutralized atomic bunches entered the laser-atom interaction region. They were collinearly overlapped with the laser pulses and synchronized with the duty cycle of the laser system. The ionization scheme used in the experiment is shown in Fig. 1 and probes the same transition from previous work on gallium isotopes [2, 12, 28]. 417.2-nm laser light was produced by frequency-doubling light from an M-Squared SolsTiS continuous-wave Ti:Sa laser (pumped by a Sprout G-18 18W continuous-wave DPSS laser), using an ECD-X frequency doubler. The chopping of this light was performed with the Pockels cell setup as detailed in Ref. [29]. A 35-m multimode optical fibre was used to transport the 417.2-nm laser light from the CRIS laser laboratory to the CRIS beam-line. The 639.7-nm light was produced from a Spectron pulsed dye laser, pumped with 532-nm light from a Litron LPY-601 Nd:YAG laser. This laser system had two laser heads which could be controlled independently. The other laser head produced the 1064-nm light required for the ionization step. The interaction region was maintained at 2×10^{-8} mbar to minimize the background due to nonresonant collisional ionization with gas molecules and isobaric contamination (such as rubidium). After resonant excitation and laser ionization, the ions were detected by deflecting them onto a Hamamatsu F4655-12 positive-ion microchannel plate (MCP) detector biased at -2250 V.

III. ANALYSIS AND RESULTS

Previous use of the target-ion-source unit with direct impact of the protons on the target resulted in large beams of isobaric rubidium and strontium contamination. Produced via proton-induced fragmentation, long-lived and stable isotopes of these elements had accumulated in the target matrix and were released and surface ionized throughout the experiment. This prevented measurements of exotic gallium isotopes past ${}^{82}\text{Ga}$ from being performed. Despite relatively high count rates of ${}^{65,67}\text{Ga}$ in Fig. 2, considerable background rates are also observed. This contamination is suspected to be stable titanium compounds [31] and

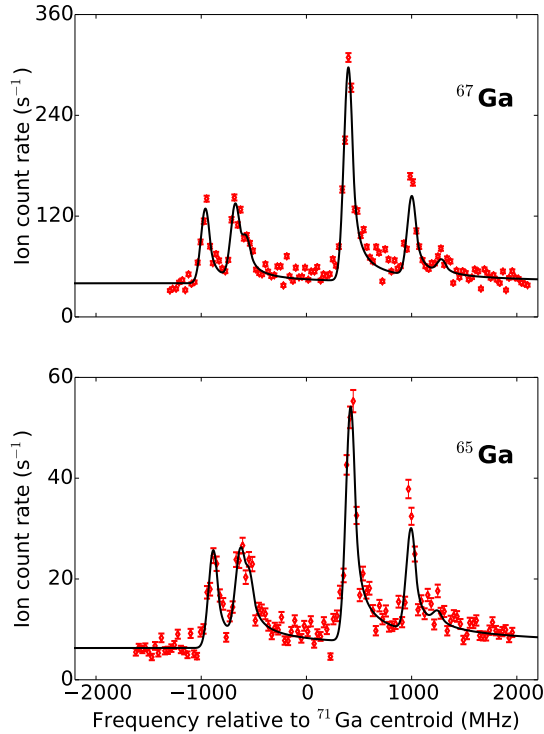


FIG. 2. (Color online) Example hyperfine spectra for ${}^{65,67}\text{Ga}$, using the $4p\ 2P_{3/2} \rightarrow 5s\ 2S_{1/2}$ transition. The solid lines are the best fit using an asymmetric Gaussian profile, assuming a spin assignment of $I = 3/2^-$.

TABLE I. The $A(4p\ ^2P_{3/2})$ and $A(5s\ ^2S_{1/2})$ factors and magnetic-dipole moments for the gallium isotopes studied in this experiment. Statistical uncertainties are enclosed with parentheses.

A	I	$A(4p\ ^2P_{3/2})$ (MHz)		$A(5s\ ^2S_{1/2})$ (MHz)		μ (μ_N)		
		This work	Cheal <i>et al.</i> ^a	This work	Cheal <i>et al.</i> ^a	This work	Cheal <i>et al.</i> ^a	Ref. [30]
65	3/2	+166.7(6)	—	+940.3(13)	—	+1.774(3)	—	—
67	3/2	+174.7(38)	+175.8(10)	+978.0(61)	+979.7(25)	+1.845(12)	+1.848(5)	+1.8507(3)
69	3/2	+189.8(13)	+191.5(9)	+1067.7(18)	+1069.5(15)	+2.014(4)	+2.018(4)	+2.01659(5)
71	3/2	+242.7(6)	+242.8(7)	+1357.7(17)	+1358.2(16)	—	—	+2.56227(2)
75	3/2	+172.6(1)	+173.6(9)	+971.7(12)	+973.1(15)	+1.833(3)	+1.836(4)	—
79	3/2	+97.3(38)	+98.3(9)	+550.2(112)	+555.0(14)	+1.038(21)	+1.047(3)	—
80g	6 ^b	+1.4(7) ^c	+0.9(1) ^c	—	—	+0.061(29) ^d	+0.036(4) ^d	—
80m	3 ^b	-68.0(2) ^c	-67.5(1) ^c	—	—	-1.435(5) ^d	-1.425(5) ^d	—
81	5/2	+98.6(10)	+98.9(4)	+555.7(26)	+555.6(13)	+1.747(8)	+1.747(5)	—
82	2 ^b	—	—	+178.7(49) ^c	+182.5(14) ^c	+0.449(12)	+0.459(4)	—

^a Values taken from Refs. [2, 12, 28].

^b Spins based on suggested assignments for ^{80}Ga [2] and ^{82}Ga [28].

^c $A(5s\ ^2S_{1/2})/A(4p\ ^2P_{3/2})$ constrained to +5.592(9) [12].

^d Magnetic moment calculated using $A(4p\ ^2P_{3/2})$ values.

has already been reported in previous studies around the same mass region at the ISOLDE facility [32].

The extraction of the ground-state properties from the hyperfine spectra for each isotope was performed with a χ^2 -minimization routine [33]. High-frequency tails have been observed in all spectra analysed in this experiment, resulting from AC Stark shift effects induced by the high power of the ionization laser. Although recent studies have suggested these distortions can be removed by delaying the ionization laser light [34], the short lifetime of the $5s\ ^2S_{1/2}$ state ($t_{1/2} = 5.7(6)$ ns [35]) did not permit this. Therefore, asymmetric fitting profiles defined by a Gaussian function with a power-law high-frequency tail were used (colloquially referred to as the Crystal Ball function) [36, 37]. The analysis of the data was performed with no restrictions on the A and B hyperfine parameters and the intensities of the hyperfine peaks, apart from the case of $^{80g,m}\text{Ga}$. Spin assignments were deduced by

TABLE II. The $B(4p\ ^2P_{3/2})$ factors and electric-quadrupole moments for the gallium isotopes studied in this experiment. Statistical uncertainties are enclosed with parentheses.

A	I	$B(4p\ ^2P_{3/2})$ (MHz)		Q_s (efm ²)		
		This work	Cheal <i>et al.</i> ^a	This work	Cheal <i>et al.</i> ^a	Ref. [38]
65	3/2	+75(1)	—	+20.7(11)	—	—
67	3/2	+71(4)	+73(4)	+19.5(15)	+19.8(16)	+19.7(2)
69	3/2	+61(8)	+63(2)	+16.7(24)	+17.1(11)	+17.1(2)
71	3/2	+38(3)	+39(2)	—	—	+10.7(1)
75	3/2	-107(2)	-105(1)	-29.5(16)	-28.5(17)	—
79	3/2	+49(12)	+58(2)	+13.4(33)	+15.8(10)	—
80g	6 ^b	+186(7)	+174(3)	+50.7(35)	+47.8(27)	—
80m	3 ^b	+132(3)	+137(3)	+35.8(23)	+37.5(21)	—
81	5/2	-12(5)	-18(3)	-3.3(13)	-4.8(8)	—
82	2 ^b	+74(29)	+72(3)	+20.0(80)	+19.7(13)	—

^a Values taken from Refs. [2, 12, 28].^b Spins based on suggested assignments for ${}^{80}\text{Ga}$ [2] and ${}^{82}\text{Ga}$ [28].

comparing ratios of the resulting A -hyperfine factors for a particular spin assignment to $A(5s\ ^2S_{1/2})/A(4p\ ^2P_{3/2}) = +5.592(9)$, arising from the weighted mean of ratios for gallium isotopes measured in Ref. [12]. Magnetic-dipole and electric-quadrupole moments were extracted from the known ratios $\mu = \mu_{\text{ref}}IA/I_{\text{ref}}A_{\text{ref}}$ and $Q_s = BQ_{\text{ref}}/B_{\text{ref}}$ and are displayed in Tables I and II, respectively. Measured observables were determined relative to ${}^{71}\text{Ga}$, using $A_{\text{ref}}(5s\ ^2S_{1/2}) = +1358.2(16)$ MHz [12], $\mu_{\text{ref}} = +2.56227(2)$ μ_N [30], $B_{\text{ref}}(4p\ ^2P_{3/2}) = +39(2)$ MHz [12], and $Q_{\text{ref}} = +10.7(1)$ efm² [39]. The low Z of gallium meant that sensitivity to the hyperfine anomaly could be neglected [40].

Isotope shifts were extracted by comparing the centroid frequency of a given spectrum to the nearest ${}^{71}\text{Ga}$ spectrum taken in time. These values are presented in the second column of Table III and compared with values from previous studies in the fourth column [4]. Additionally, Fig. 3 plots these values from both studies against each other. The systematic offset between the two data sets is highlighted by the deviation of the green dotted and blue solid lines. It can be attributed to an offset in the ISCOOL acceleration voltage, which is

TABLE III. Isotope-shift values for the isotopes studied in this work. Statistical and systematic uncertainties are enclosed in parentheses and square brackets respectively. See text and Fig. 3 for further details.

A	$\delta\nu^{71,A}$ (MHz)		
	This work	Corrected values	Procter <i>et al.</i> [4]
65	+101(5)	+106(6)[0.8]	—
67	+74(11)	+77(12)[0.6]	—
69	+33(35)	+34(36)[0.3]	+40(4)
71	0	0	0
75	−44(4)	−46(4)[0.4]	−45(2)
79	−173(11)	−182(12)[1.4]	−186(2)
80g	−230(4)	−241(4)[1.8]	−239(4)
80m	−224(4)	−235(5)[1.8]	−232(3)
81	−252(7)	−264(8)[2.0]	−272(2)
82	−207(4)	−217(5)[1.7]	−222(9)

known to vary between experiments at the ISOLDE facility [4, 41, 42]. In order to directly compare between the two data sets, isotope shifts obtained in this study were modified by applying a correction factor given by the gradient difference between the two lines in Fig. 3, given by

$$\delta\nu_{Mod}^{71,A} = 1.049(8)\delta\nu_{CRIS}^{71,A}, \quad (1)$$

where $\delta\nu_{Mod}^{71,A}$ represents modified isotope shifts obtained in this study, which are presented in the third column of Table III. This modification benchmarks these values to previous literature [4] and, in particular for $^{65,67}\text{Ga}$, makes them directly comparable to the rest of the isotope shifts that have already been obtained across the gallium chain. The associated error represents an additional systematic uncertainty and is quoted separately for the modified isotope shifts given in Table III. Changes in the mean-square charge radii were calculated from the modified isotope shifts by using the equation

$$\delta\nu^{AA'} = M_{417} \frac{A' - A}{AA'} + F_{417} \delta\langle r^2 \rangle^{AA'}, \quad (2)$$

where A and A' are the atomic masses assigned to the reference and measured isotopes re-

spectively [43]. The parameters $M_{417} = -211.4(210)$ GHz amu and $F_{417} = 0.40(6)$ GHz/fm² represent the atomic factors and contain all the optical dependence for the 417.2-nm transition [4]. Reported hyperfine coefficients and isotope shift values for $^{67,69,75,79-82}\text{Ga}$ in Tables I, II, and III are consistent with tabulated literature values [2, 4, 12, 28]. Hyperfine parameters for $A(^2P_{3/2})$, $A(^2S_{1/2})$, $B(^2P_{3/2})$, and $\delta\nu_{CRIS}^{71,A}$ were determined from a weighted mean of values obtained from the analysis of individual hyperfine spectra. Statistical uncertainties were determined from a weighted standard deviation of the individual values.

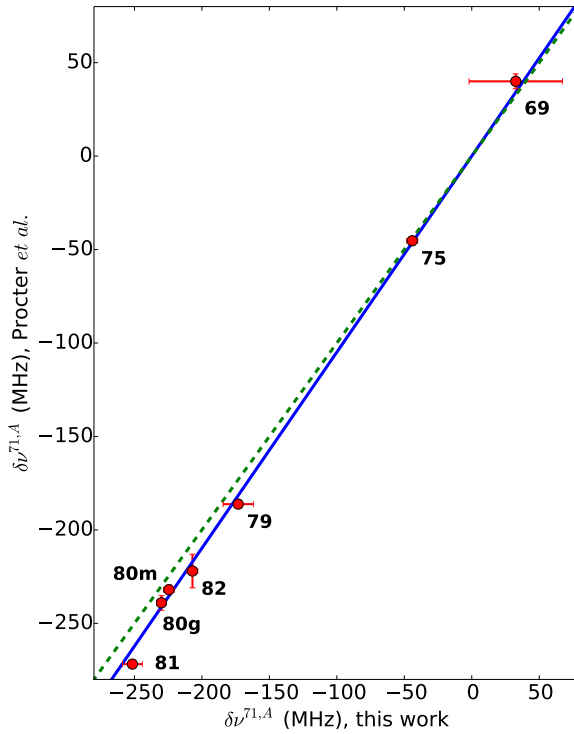


FIG. 3. (Color online) Isotope shift values from literature [4] plotted against this work. The green-dotted line represents consistency between the two data sets and the blue solid line represents the current relation between the two. Each data point is labeled according to the mass number involved.

IV. DISCUSSION

A. Electromagnetic moments for $28 \leq N \leq 40$

An analysis of the hyperfine A factors for ^{65}Ga revealed ratios of $+5.641(32)$ and $+5.686(37)$ for $I = 3/2^-$ and $I = 5/2^-$ assignments. This results in standard deviations of 1σ and 2σ , respectively, from $+5.592(9)$ [12]. Therefore, $I = 3/2^-$ is assigned for ^{65}Ga and confirms the ground-state spin assignment already made for this isotope [44]. Similar analyses for $^{67,69}\text{Ga}$ also confirm the ground-state spins of $I = 3/2^-$ as reported in Ref. [12].

Shell-model calculations of the nuclear moments for the lowest-lying $3/2^-$ states for $^{59-71}\text{Ga}$ were performed using the KSHELL code [45] with two effective interactions: GXPF1 [46] and JUN45 [47]. The model spaces used for each interaction are displayed in Fig. 4.

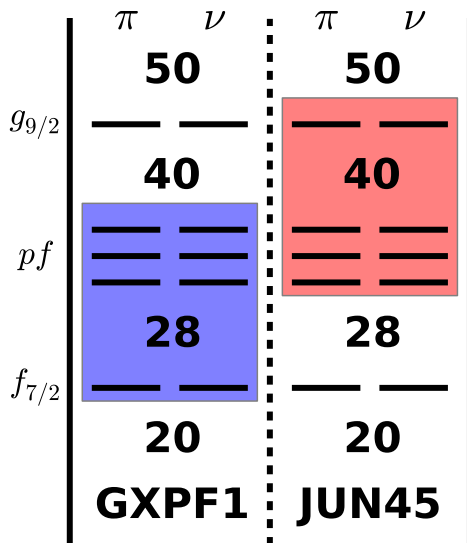


FIG. 4. (Color online) A schematic representation of the model spaces involved for the GXPF1 and JUN45 interactions, shaded in blue and red respectively.

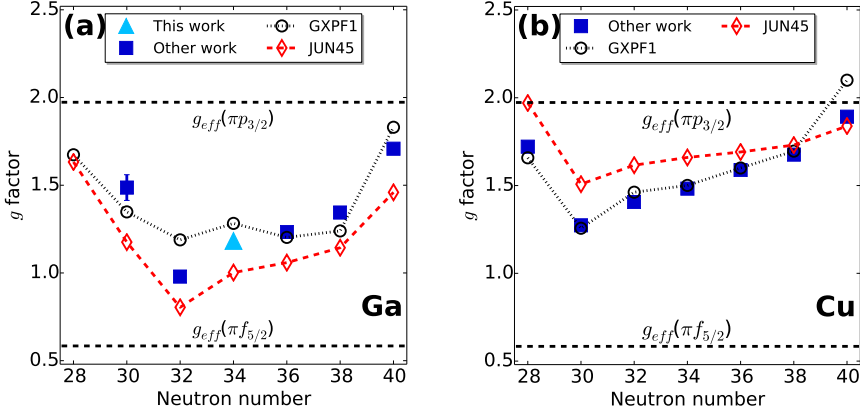


FIG. 5. (Color online) (a) The extracted g factor for ${}^{65}\text{Ga}$ (light-blue triangle), alongside tabulated values across the gallium isotope chain from Refs. [4, 30, 48] (dark-blue squares). Apart from ${}^{61}\text{Ga}$, the error bars are smaller than the marker size. (b) g factors for the odd- A copper isotopes in the same mass range [15] (dark-blue squares). Values are compared to GXPF1 (black circles) and JUN45 (red diamonds) calculations. Effective g -factor values for the $\pi p_{3/2}$ and $\pi f_{5/2}$ orbitals are illustrated with dashed lines.

GXPF1 calculations are performed in a full pf model space with respect to the ${}^{40}\text{Ca}$ core. The values reported in this work are performed with up to 2p-2h excitations across the $N, Z = 28$ shell closures for the neutrons and protons, respectively. Calculations allowing up to 4p-4h excitations revealed that a convergence is reached at the 2p-2h level. This interaction has already been used to interpret the moments for the neutron-deficient odd- A copper isotopes down to ${}^{57}\text{Cu}$ [15, 17], in addition to the quadrupole moments for the odd- A manganese isotopes in the same mass region [14].

The JUN45 calculations are performed starting from a ${}^{56}\text{Ni}$ core (see Fig. 4), including the upper pf orbits and the $g_{9/2}$ orbit as a valence space for both protons and neutrons. Calculations with this interaction were performed previously, in order to compare with moment measurements of ${}^{63,67,69,71}\text{Ga}$ [4, 33]. In this article, we extend this study by including ${}^{59,61,65}\text{Ga}$. The following effective g factors and effective charges were used for GXPF1: $g_s^{eff} = 0.9g_s^{free}$, $g_l^{eff} = +1.1, -0.1$ and $e_p^{eff} = 1.5e$, $e_n^{eff} = 0.5e$, respectively [46]. For JUN45,

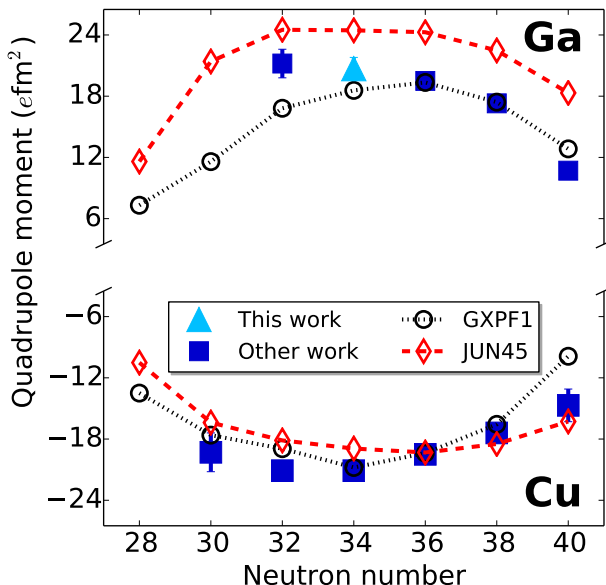


FIG. 6. (Color online) The extracted electric quadrupole moment for ^{65}Ga (light-blue triangle), alongside literature values for the gallium [4, 38] and copper [17, 49] isotope chains (dark-blue squares). Values are compared to GXPF1 (black circles) and JUN45 (red diamonds) calculations.

the values $g_s^{eff} = 0.7g_s^{free}$, $g_l^{eff} = +1.0$, 0 and $e_p^{eff} = 1.5e$, $e_n^{eff} = 1.1e$ were used [47].

The g factors for $^{61-71}\text{Ga}$ are presented in Fig. 5(a). Values for the Cu isotope chain are presented alongside in Fig. 5(b) for comparison. With only one proton outside $Z = 28$, the GXPF1 calculations reproduce the Cu magnetic moments very well, due to the inclusion of excitations across the $N, Z = 28$ shell closures. Indeed with the JUN45 interaction, in which these excitations are not included in the model space, the Cu magnetic moment trends are less well reproduced. For the Ga isotopes however, the agreement of both GXPF1 and JUN45 interactions is similar.

Quadrupole moment values for the same isotopes are presented in Fig. 6. An increase towards the midshell at $N = 34$ (between $N = 28$ and the $N = 40$ subshell gap) confirms the increasing presence of neutron correlations, similar to the trend seen in the copper

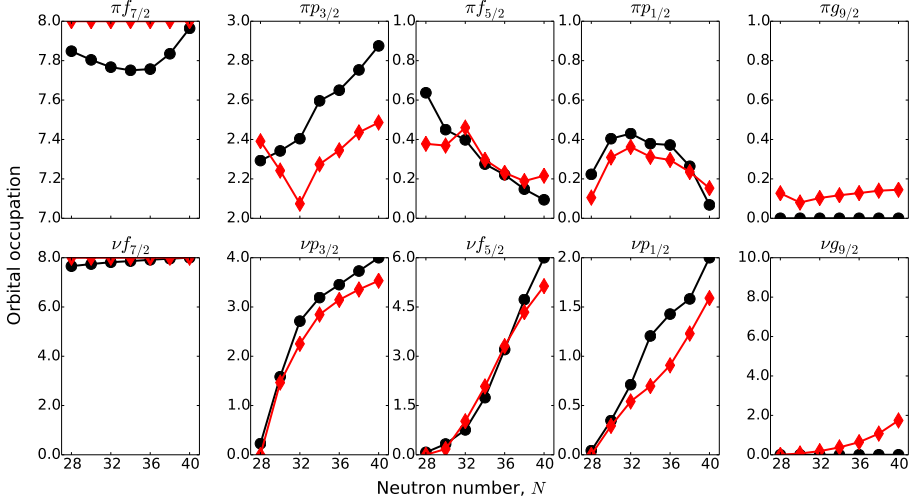


FIG. 7. (Color online) Proton and neutron orbital occupations for odd- A ${}^{59-71}\text{Ga}$, across the combined orbital space for the GXPF1 (black circles) and JUN45 (red diamonds) interactions. Proton occupancies (top) are viewed with respect to the occupancy of a single proton, while neutron occupancies (bottom) are viewed with respect to the total occupancy for a given orbital.

quadrupole moments [17]. Comparable parabolic trends can be seen in the experimental $B(E2)$ values between $N = 28$ and 50 for the nickel isotopes [50], and between $N = 50$ and 82 for the tin isotopes [51]. It is also noted that the quadrupole moments of the odd- A Ga isotopes and those of the odd- A Cu isotopes have a similar magnitude, but an opposite sign. This reflects the rather pure nature of these configurations: for a pure $\pi p_{3/2}^n$ configuration, one expects that $Q_{sp}(\pi p_{3/2}^1, \text{Cu}) = -Q_{sp}(\pi p_{3/2}^3, \text{Ga})$ [52].

Figure 7 illustrates the changes in the proton and neutron orbital occupations in the odd- A Ga wave functions, across the combined GXPF1 and JUN45 model spaces displayed in Fig. 4. The striking similarity between the trends in the JUN45 $\pi p_{3/2}$ orbital occupancies and the magnetic moment values described in Fig. 5 suggests that contributions from this orbital are important to describe the magnetic moments between $N = 28$ and 40. While the ${}^{59-71}\text{Ga}$ moments predicted by JUN45 correspond to mixed $\pi p_{3/2}/\pi f_{5/2}$ configurations over the full range (due to the scattering of protons in the upper pf -orbitals), the $\pi p_{3/2}$ occupancy for GXPF1 increases towards $N = 40$. This illustrates how blocking the neutron orbits

above $N = 40$ leads to reduced proton excitations across $Z = 28$ as the neutron shell is being filled. Furthermore, although proton and neutron excitations from the $f_{7/2}$ orbital are allowed in this case, this does not improve the reproduction of the experimental moment values.

B. Changes in the mean-square charge radii for $^{65,67}\text{Ga}$

The changes in mean-square charge radii for $^{65,67}\text{Ga}$ are given in Table IV and displayed graphically alongside literature values in Fig. 8. For this discussion, neutron numbers are used for the charge-radii superscripts. Uncertainties quoted in Table IV originate from the statistical and systematic errors quoted in Table III, plus an additional systematic uncertainty from the F_{417} and M_{417} atomic factors. The odd-even staggering (OES) was investigated by calculating the $D(N; \delta \langle r^2 \rangle^{40,N})$ parameter [53, 54], given by

$$D(N; \delta \langle r^2 \rangle^{40,N}) = (-1)^N \left[\delta \langle r^2 \rangle^{40,N} - \frac{\delta \langle r^2 \rangle^{40,(N-1)} + \delta \langle r^2 \rangle^{40,(N+1)}}{2} \right], \quad (3)$$

and is also shown in Fig. 8. A normal odd-even staggering corresponds to positive D values, corresponding to odd-odd Ga isotopes having a slightly smaller charge radius than their odd-even neighbors. This is due to the odd neutron in even- N isotopes blocking the scattering of boson 0^+ pairs over several orbits from its own orbital [55]. Values with $D(N; \delta \langle r^2 \rangle^{40,N}) < 0$ can be attributed to a reversal in OES. This has been observed in the $N \approx 40$ region not only with gallium [4], but also in the krypton and strontium chains [56]. It is also observed in the $N = 134$ region in the francium and radium chains, in which it coincides with a region of octupole deformation [54, 57]. The reversal at $N = 50$ is caused by the dramatic increase

TABLE IV. Changes in the mean-square charge radii for $^{65,67}\text{Ga}$ ($N = 34, 36$), using the modified experimental isotope shift values in Table III. The statistical, modified-systematic and atomic-systematic uncertainties are enclosed within parentheses, square brackets, and curly brackets, respectively.

N	$\delta \langle r^2 \rangle^{40,N}$ (fm ²)
34	-0.422(14)[2]{93}
36	-0.252(30)[2]{58}

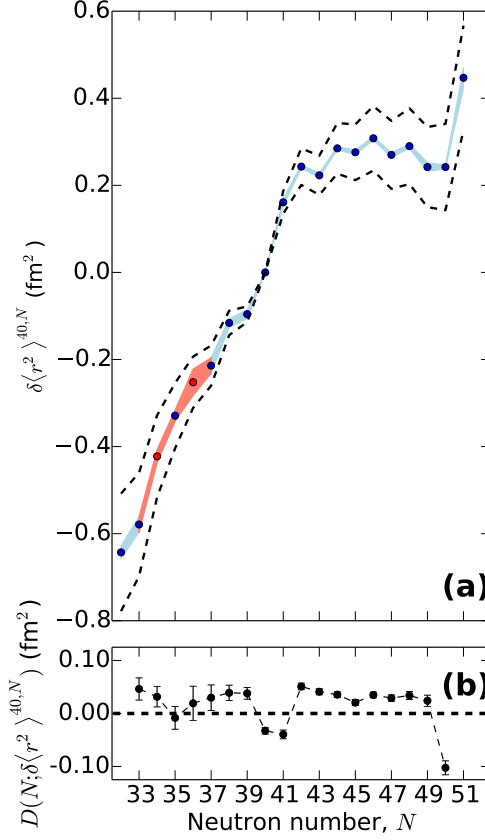


FIG. 8. (Color online) (a) Changes in the mean-square charge radii across the gallium isotope chain. Red circles represent new measurements for $^{65,67}\text{Ga}$ ($N = 34, 36$) and blue circles denote literature values [4]. Statistical and atomic systematic uncertainty bounds are given by the shaded areas and black-dotted lines respectively. (b) Corresponding $D(N; \delta\langle r^2 \rangle^{40,N})$ values representing the odd-even staggering, calculated from Eq. (3).

in the charge-radius value of ^{82}Ga at $N = 51$, the signature of a shell closure crossing [4, 5]. Further investigation with greater isotope shift sensitivity is also required to assert odd-even staggering characteristics at $N = 35$.

V. CONCLUSION

Gallium isotopes were studied at the CRIS experiment at ISOLDE, with the use of a three-step laser ionization scheme. The electromagnetic moments of ^{65}Ga and the changes in the mean-square charge radii for $^{65,67}\text{Ga}$ are reported. A comparison of the moments for $28 \leq N \leq 40$ with shell-model calculations suggest that, as protons occupy the upper pf orbits, calculations starting from a ^{56}Ni core reproduce the moments equally well as calculations that allow proton and neutron excitations across $N, Z = 28$. The inclusion of the $\nu g_{9/2}$ neutron orbit is only important for isotopes near $N = 40$. For the mean-square charge-radii values, an inverted odd-even staggering is observed around $N = 40$, although an explanation for this has not yet been proposed. A small inversion is observed also at $N = 35$; however, a remeasurement of the isotope shifts with better voltage precision is suggested to assert OES inversion in this region.

VI. ACKNOWLEDGEMENTS

We acknowledge the support of the ISOLDE Collaboration and technical teams. This work was supported by the ERC Consolidator Grant No.648631; the IUAP-Belgian State Belgian Science Policy (BRIX network P7/12), FWO-Vlaanderen (Belgium) and GOA 15/010 from KU Leuven; the Science and Technology Facilities Council Consolidated Grant No.ST/F012071/1, Continuation Grant No.ST/J000159/1 and Ernest Rutherford Grant No.ST/L002868/1; and the European Union's Seventh Framework Programme for Research and Technological Development under Grant Agreements No. 262010 (ENSAR), No. 267194 (COFUND), and No. 289191 (LA³NET). T.E.C. was supported by the STFC Ernest Rutherford Fellowship No.ST/J004189/1. We acknowledge the financial aid from the Ed Schneiderman Fund at New York University.

-
- [1] X. F. Yang, C. Wraith, L. Xie, C. Babcock, J. Billowes, M. L. Bissell, K. Blaum, B. Cheal, K. T. Flanagan, R. F. Garcia Ruiz, W. Gins, C. Gorges, L. K. Grob, H. Heylen, S. Kaufmann, M. Kowalska, J. Kraemer, S. Malbrunot-Ettenauer, R. Neugart, G. Neyens, W. Nörtershäuser, J. Papuga, R. Sánchez, and D. T. Yordanov, Phys. Rev. Lett. **116**, 182502 (2016).

- [2] B. Cheal, J. Billowes, M. L. Bissell, K. Blaum, F. C. Charlwood, K. T. Flanagan, D. H. Forest, S. Fritzsche, C. Geppert, A. Jokinen, M. Kowalska, A. Krieger, J. Krämer, E. Mané, I. D. Moore, R. Neugart, G. Neyens, W. Nörtershäuser, M. M. Rajabali, M. Schug, H. H. Stroke, P. Vingerhoets, D. T. Yordanov, and M. Záková, *Phys. Rev. C* **82**, 051302(R) (2010).
- [3] A. Gottardo, D. Verney, C. Delafosse, F. Ibrahim, B. Roussière, C. Sotty, S. Roccia, C. Andreoiu, C. Costache, M. C. Delattre, I. Deloncle, A. Etilé, S. Franchoo, C. Gaulard, J. Guillet, M. Lebois, M. MacCormick, N. Marginean, R. Marginean, I. Matea, C. Mihai, I. Mitu, L. Olivier, C. Portail, L. Qi, L. Stan, D. Testov, J. Wilson, and D. T. Yordanov, *Phys. Rev. Lett.* **116**, 182501 (2016).
- [4] T. J. Procter, J. Billowes, M. L. Bissell, K. Blaum, F. C. Charlwood, B. Cheal, K. T. Flanagan, D. H. Forest, S. Fritzsche, C. Geppert, H. Heylen, M. Kowalska, K. Kreim, A. Krieger, J. Krämer, K. M. Lynch, E. Mané, I. D. Moore, R. Neugart, G. Neyens, W. Nörtershäuser, J. Papuga, M. M. Rajabali, H. H. Stroke, P. Vingerhoets, D. T. Yordanov, and M. Záková, *Phys. Rev. C* **86**, 034329 (2012).
- [5] P. Campbell, I. D. Moore, and M. R. Pearson, *Prog. Par. Nucl. Phys.* **86**, 127 (2016).
- [6] K. Sieja and F. Nowacki, *Phys. Rev. C* **85**, 051301(R) (2012).
- [7] Y. Tsunoda, T. Otsuka, N. Shimizu, M. Honma, and Y. Utsuno, *Phys. Rev. C* **89**, 031301(R) (2014).
- [8] G. Hagen, G. R. Jansen, and T. Papenbrock, *Phys. Rev. Lett.* **117**, 172501 (2016).
- [9] F. Nowacki, A. Poves, E. Caurier, and B. Bounthong, *Phys. Rev. Lett.* **117**, 272501 (2016).
- [10] S. Franchoo, M. Huyse, K. Kruglov, Y. Kudryavtsev, W. F. Mueller, R. Raabe, I. Reusen, P. Van Duppen, J. Van Roosbroeck, L. Vermeeren, A. Wöhr, H. Grawe, K. L. Kratz, B. Pfeiffer, and W. B. Walters, *Phys. Rev. C* **64**, 054308 (2001).
- [11] K. T. Flanagan, P. Vingerhoets, M. Avgoulea, J. Billowes, M. L. Bissell, K. Blaum, B. Cheal, M. De Rydt, V. N. Fedosseev, D. H. Forest, C. Geppert, U. Köster, M. Kowalska, J. Krämer, K. L. Kratz, A. Krieger, E. Mané, B. A. Marsh, T. Materna, L. Mathieu, P. L. Molkanov, R. Neugart, G. Neyens, W. Nörtershäuser, M. D. Seliverstov, O. Serot, M. Schug, M. A. Sjoedin, J. R. Stone, N. J. Stone, H. H. Stroke, G. Tungate, D. T. Yordanov, and Y. M. Volkov, *Phys. Rev. Lett.* **103**, 142501 (2009).
- [12] B. Cheal, E. Mané, J. Billowes, M. L. Bissell, K. Blaum, B. A. Brown, F. C. Charlwood, K. T. Flanagan, D. H. Forest, C. Geppert, M. Honma, A. Jokinen, M. Kowalska, A. Krieger,

- J. Krämer, I. D. Moore, R. Neugart, G. Neyens, W. Nörtershäuser, M. Schug, H. H. Stroke, P. Vingerhoets, D. T. Yordanov, and M. Záková, *Phys. Rev. Lett.* **104**, 252502 (2010).
- [13] C. J. Prokop, B. P. Crider, S. N. Liddick, A. D. Ayangeakaa, M. P. Carpenter, J. J. Carroll, J. Chen, C. J. Chiara, H. M. David, A. C. Dombos, S. Go, J. Harker, R. V. F. Janssens, N. Larson, T. Lauritsen, R. Lewis, S. J. Quinn, F. Recchia, D. Seweryniak, A. Spyrou, S. Suchyta, W. B. Walters, and S. Zhu, *Phys. Rev. C* **92**, 061302(R) (2015).
- [14] B. Babcock, H. Heylen, *et al.*, *Phys. Lett. B* **750**, 176 (2016).
- [15] 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, *Phys. Rev. Lett.* **103**, 102501 (2011).
- [16] T. Otsuka, M. Honma, and T. Mizusaki, *Phys. Rev. Lett.* **81**, 1588 (1998).
- [17] P. Vingerhoets, K. T. Flanagan, *et al.*, *Phys. Lett. B* **703**, 34 (2011).
- [18] E. Kugler, *Hyperfine Interact.* **129**, 23 (2000).
- [19] A. Herlert, *Nucl. Phys. News* **20**, 5 (2010).
- [20] A. Gottberg *et al.*, *Nucl. Instrum. Methods B* **336**, 143 (2014).
- [21] B. A. Marsh *et al.*, *Nucl. Instrum. Methods B* **317**, 550 (2013).
- [22] H. Frånberg *et al.*, *Nucl. Instrum. Methods B* **266**, 4502 (2008).
- [23] E. Mané *et al.*, *Eur. Phys. J. A* **42**, 503 (2009).
- [24] T. E. Cocolios *et al.*, *Nucl. Instrum. Methods B* **317**, 565 (2013).
- [25] K. M. Lynch, J. Billowes, M. L. Bissell, I. Budinčević, T. E. Cocolios, R. P. De Groote, S. De Schepper, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt, *Phys. Rev. X* **4**, 011055 (2014).
- [26] T. E. Cocolios, R. P. de Groote, *et al.*, *Nucl. Instrum. Methods B* **376**, 284 (2016).
- [27] D. Rapp and W. E. Francis, *J. Chem. Phys.* **37**, 2631 (1962).
- [28] B. Cheal *et al.*, *J. Phys.: Conf. Ser.* **381**, 012071 (2012).
- [29] 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, *Phys. Rev. Lett.* **115**, 132501 (2015).
- [30] N. J. Stone, *At. Data Nucl. Data Tables* **90**, 75 (2005).

- [31] F. Weinholtz and ISOLTRAP, (private communication).
- [32] C. Guénaut, G. Audi, D. Beck, K. Blaum, G. Bollen, P. Delahaye, F. Herfurth, A. Kellerbauer, H. J. Kluge, J. Libert, D. Lunney, S. Schwarz, L. Schweikhard, and C. Yazidjian, *Phys. Rev. C* **75**, 044303 (2007).
- [33] B. Cheal and K. T. Flanagan, *J. Phys. G: Nucl. Phys.* **37**, 113101 (2010).
- [34] R. P. de Groote, M. Verlinde, V. Sonnenschein, K. T. Flanagan, I. Moore, and G. Neyens, *Phys. Rev. A* **95**, 032502 (2017).
- [35] U. I. Safronova *et al.*, *J. Phys. B: At. Mol. Opt. Phys.* **39**, 749 (2006).
- [36] J. Gaiser, *Ph.D thesis*, Ph.D. thesis, Stanford University, Stanford, California, USA (1982), SLAC-0255 (unpublished).
- [37] T. Skwarnicki, *Ph.D thesis*, Ph.D. thesis, Institute of Nuclear Physics, Cracow, Poland (1986), DESY-F31-86-02 (unpublished).
- [38] N. J. Stone, *At. Data Nucl. Data Tables* **111**, 1 (2005).
- [39] P. Pyykkö, *Mol. Phys.* **106**, 16 (2008).
- [40] H. H. Stroke, R. J. Blin-Stoyle, and V. Jaccarino, *Phys. Rev.* **123**, 1326 (1961).
- [41] P. Vingerhoets, *Ph.D thesis*, Ph.D. thesis, KU Leuven, Belgium (unpublished) (2011).
- [42] J. Papuga, *Ph.D thesis*, Ph.D. thesis, KU Leuven, Belgium (unpublished) (2015).
- [43] M. Wang *et al.*, *Chin. Phys. C* **36**, 1603 (2012).
- [44] I. Dankó, D. Sohler, Z. Dombrádi, S. Brant, V. Krstić, J. Cederkäll, M. Lipoglavšek, M. Palacz, J. Persson, A. Atac, C. Fahlander, H. Grawe, A. Johnson, A. Kerek, W. Klamra, J. Kownacki, A. Likar, L. O. Norlin, J. Nyberg, V. Paar, R. Schubart, D. Seweryniak, D. Vretenar, G. de Angelis, P. Bednarczyk, D. Foltescu, D. Jerrestam, S. Juutinen, E. Mäkelä, B. M. Nyakó, M. de Poli, H. A. Roth, T. Shizuma, Ö. Skeppstedt, G. Sletten, and S. Törmänen, *Phys. Rev. C* **59**, 1956 (1999).
- [45] N. Shimizu, , arXiv:1310.5431 (2013).
- [46] M. Honma, T. Otsuka, B. A. Brown, and T. Mizusaki, *Phys. Rev. C* **69**, 034335 (2004).
- [47] M. Honma, T. Otsuka, T. Mizusaki, and M. Hjorth-Jensen, *Phys. Rev. C* **80**, 064323 (2009).
- [48] L. Weissman, J. Cederkall, J. Äystö, H. Fynbo, L. Fraile, V. Fedosseyev, S. Franchoo, A. Jokinen, U. Köster, G. Martínez-Pinedo, T. Nilsson, M. Oinonen, K. Peräjärvi, M. D. Seliverstov, and the ISOLDE Collaboration, *Phys. Rev. C* **65**, 044321 (2002).

- [49] P. Vingerhoets, K. T. Flanagan, M. Avgoulea, J. Billowes, M. L. Bissell, K. Blaum, B. A. Brown, B. Cheal, M. De Rydt, D. H. Forest, C. Geppert, M. Honma, M. Kowalska, J. Krämer, A. Krieger, E. Mané, R. Neugart, G. Neyens, W. Nörtershäuser, T. Otsuka, M. Schug, H. H. Stroke, G. Tungate, and D. T. Yordanov, *Phys. Rev. C* **82**, 064311 (2010).
- [50] J. M. Allmond, B. A. Brown, A. E. Stuchbery, A. Galindo-Uribarri, E. Padilla-Rodal, D. C. Radford, J. C. Batchelder, M. E. Howard, J. F. Liang, B. Manning, R. L. Varner, and C. H. Yu, *Phys. Rev. C* **90**, 034309 (2014).
- [51] A. Ekström, J. Cederkäll, C. Fahlander, M. Hjorth-Jensen, F. Ames, P. A. Butler, T. Davinson, J. Eberth, F. Fincke, A. Görgen, M. Górski, D. Habs, A. M. Hurst, M. Huyse, O. Ivanov, J. Iwanicki, O. Kester, U. Köster, B. A. Marsh, J. Mierzejewski, P. Reiter, H. Scheit, D. Schwalm, S. Siem, G. Sletten, I. Stefanescu, G. M. Tveten, J. Van de Walle, P. Van Duppen, D. Voulot, N. Warr, D. Weisshaar, F. Wenander, and M. Zielińska, *Phys. Rev. Lett.* **101**, 012502 (2008).
- [52] G. Neyens, *Rep. Prog. Phys.* **66**, 633 (2003).
- [53] A. Coc *et al.*, *Nucl. Phys. A* **468**, 1 (1987).
- [54] I. Budinčević, J. Billowes, M. L. Bissell, T. E. Cocolios, R. P. de Groote, S. De Schepper, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, K. M. Lynch, B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt, *Phys. Rev. C* **90**, 014317 (2014).
- [55] D. Zawischa, *Phys. Lett. B* **155**, 309 (1985).
- [56] P. Lievens *et al.*, *Europhys. Lett.* **33**, 11 (1996).
- [57] S. A. Ahmad *et al.*, *Nucl. Phys. A* **483**, 244 (1988).

5.2 Characterisations of the reference isotope, ^{71}Ga

Qualitative statements regarding the ground-state properties of any isotope requires a thorough characterisation of a chosen reference isotope; for CRIS-setup experiments on the gallium isotope chain, ^{71}Ga was used. Throughout both IS571 experiments, a total of 23 full ^{71}Ga hyperfine spectra were collected. Weighted means for each extracted hyperfine parameter in Figure 5.2 (which are numerically summarised in Table 5.1) are not only consistent with the overall 2015 and 2017 data sets, but also consistent with previous literature values reported by the COLLAPS experiment [Che10b]. This reproducibility provides confidence for extracting the hyperfine observables of other gallium isotopes measured in-between these temporal bounds (see Chapter 5.3). Subsequent electromagnetic-moment analyses reported in the successive sections of this chapter utilise these CRIS-setup weighted means given in Table 5.1. This explains any (improved) value and uncertainty differences between the values

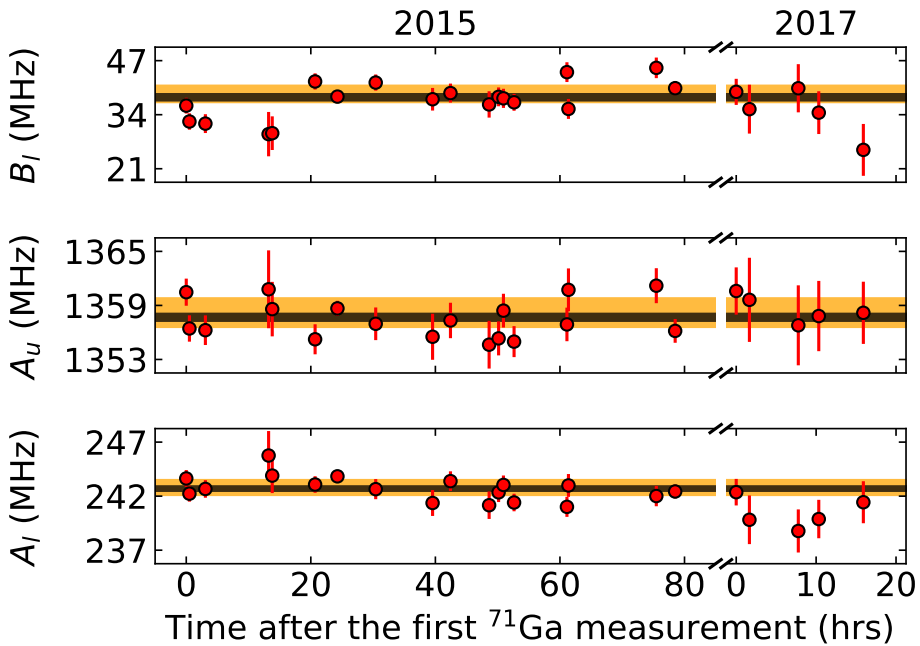


Figure 5.2: Temporal trends of extracted ^{71}Ga hyperfine parameters with respect to the start time of the first ^{71}Ga measurement, for each IS571 experiment. The shaded areas represent weighted-mean values $\pm 1\sigma$ of the entire ^{71}Ga data set presented in this thesis (black) and Ref. [Che10b] (orange).

Table 5.1: *A*- and *B*-hyperfine parameters for ⁷¹Ga, representative of weighted means from the amalgamated IS571 data set graphically displayed in Figure 5.2; the *A*-hyperfine parameter ratio is given in the right-most column. Values are compared to those obtained by the COLLAPS collaboration in the second row.

Works	<i>A</i> (MHz)		<i>B</i> (MHz)	<i>A</i> -ratio
	<i>4p</i> ² <i>P</i> _{3/2}	<i>5s</i> ² <i>S</i> _{1/2}	<i>4p</i> ² <i>P</i> _{3/2}	
CRIS	+242.7(2)	+1357.7(4)	+38(1)	+5.594(5)
Ref. [Che10b]	+242.8(7)	+1358.2(16)	+39(2)	+5.592(9)

given in this thesis, with those already reported in **Article I** which utilised ⁷¹Ga reference hyperfine parameters from Ref. [Che10b].

Centroid frequencies associated with the same ⁷¹Ga measurements used in Figure 5.2 to display their *A*- and *B*-hyperfine parameters, are shown in Figure 5.3. Despite correcting for wavemeter drifts in accordance with the frequency changes observed with the HeNe laser, there is still a considerable scatter amongst ⁷¹Ga centroid frequencies collected throughout the 2015 IS571 experiment.

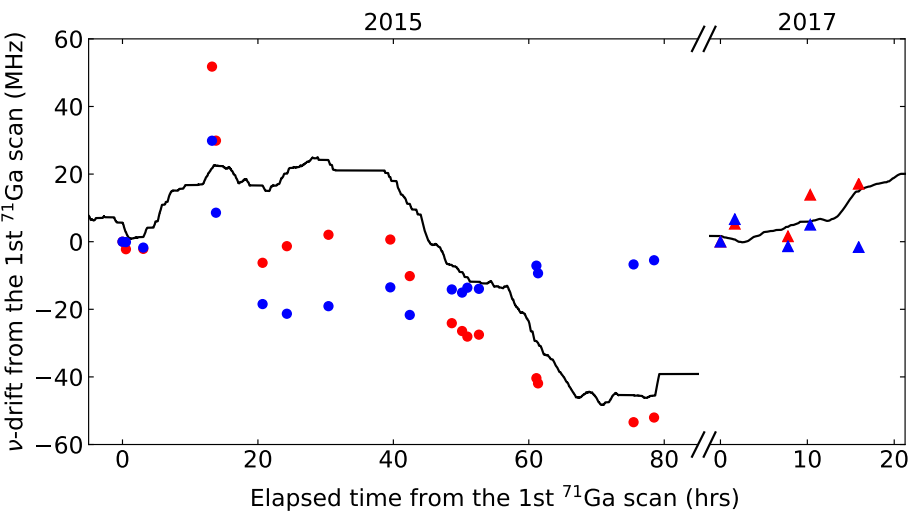


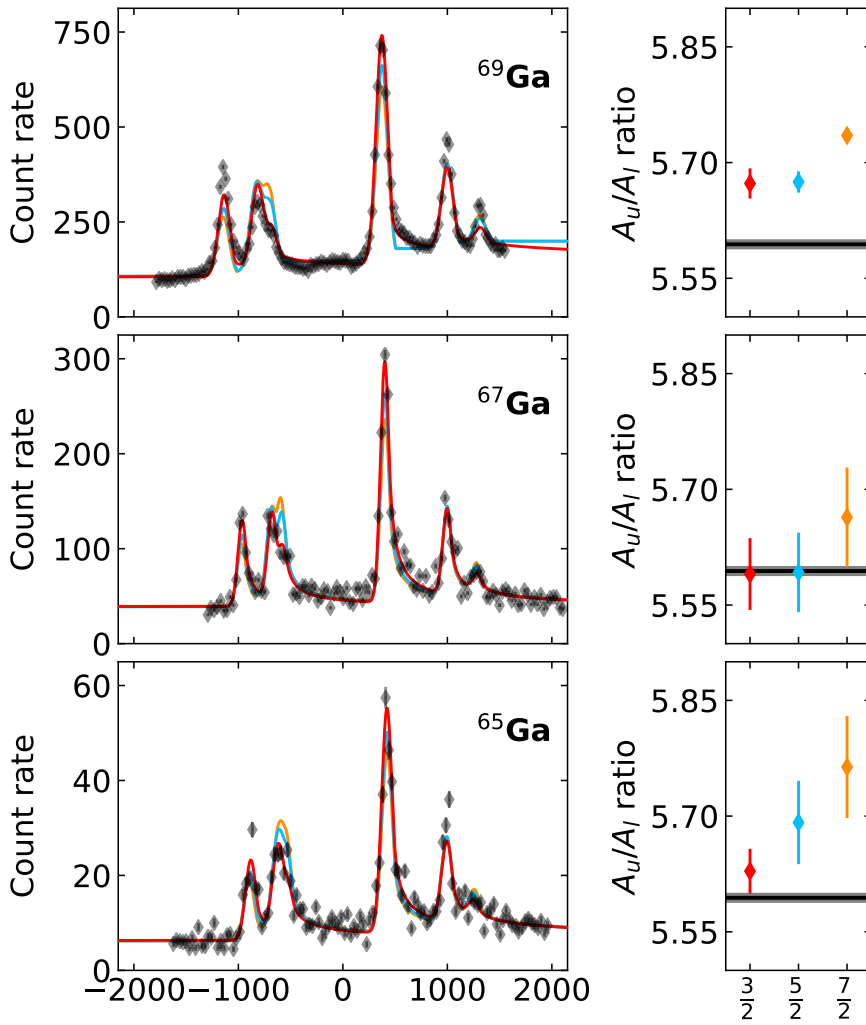
Figure 5.3: Temporal trends of extracted ⁷¹Ga centroid frequencies with respect to the start time of the first ⁷¹Ga measurement (at *t* = 0 hrs), for each IS571 experiment. Wavemeter-(un)corrected centroid frequencies are represented by the (red)/blue circles (2015) and triangles (2017) respectively. The black line represents the frequency drift as recorded by the HeNe laser.

This stresses the utmost importance of performing regular reference-isotope measurements in order to ensure the accuracy of extracted isotope-shift values. While deviations are more prominent nearer the beginning of the experiment, it is fortunate that the majority of other isotope measurements had been taken towards the end, as well as immediately before or after a hyperfine measurement of ⁷¹Ga. Isotope-shift extractions thus, involved using the closest ⁷¹Ga reference centroid frequency in time; given the limited size of the data set and lack of any additional studies to further characterise these frequency drifts, this procedure was assumed to be sufficient. Although the frequency drifts seen in the 2017 IS571 experiment featured less prominently, the same aforementioned methodology has also been applied.

5.3 Nuclear-spin-assignment analyses for ^{65,67,69}Ga

Ground-state spin analyses of ^{65,67,69}Ga have been performed using the methods outlined in Chapter 2.3.3. Although only $I = 3/2$ and $5/2$ had been investigated for each isotope in **Article I**, this thesis additionally considers the $I = 7/2$ spin assignment for completeness. This full analysis is presented in Figure 5.4, with the numerical results displayed in Table 5.2. Due to the observation of more than three hyperfine-structure resonances, the $I = 1/2$ spin assignment can be excluded for the three aforementioned gallium isotopes; see Chapter 2.3.3 for further insight.

From Table 5.2, the average reduced chi-squared values (χ_r^2) indicate an $I = 3/2$ preference for ^{65,67,69}Ga and are representative of the global behaviour for such values of each individual scan taken. Although the A -hyperfine-parameter ratio analyses also infer the same conclusion and in general rules out $I = 7/2$, some ambiguity remains with differentiating between $I = 3/2$ and $I = 5/2$. The relatively poor σ and χ_ν^2 values for ⁶⁹Ga can be attributed to non-ideal CRIS setup conditions at the beginning of the experiment in which these measurements were taken. Not only had instabilities in the frequency stepping of the 417.2-nm light been experienced, the observed hyperfine resonances also indicate the possibility of a saturated 417.2-nm transition at a time when saturation tests in Chapter 3.1.2 had not yet been performed. Correcting these effects however, are not possible because scarce quantitative information had been recorded at the beginning of this experiment. Regrettably, no further ⁶⁹Ga measurements had been taken in the remainder of the experiment which would have improved the data set. Nonetheless, assignments for ^{67,69}Ga agree with those assigned in atomic-beam resonance experiments [Ehl68; Bec48] and also from recent optical spectroscopy studies [Che10b].



Frequency relative to the centroid of ^{71}Ga (MHz) Spin

Figure 5.4: Selected hyperfine spectra of $^{65,67,69}\text{Ga}$, each fitted assuming $I = 3/2$ (red), $5/2$ (blue), and $7/2$ (orange) spin assignments. Corresponding A -hyperfine-parameter ratios extracted from the complete data set for each spin assignment are displayed on the right-hand side, compared with the CRIS-setup ratio for ^{71}Ga from Table 5.1 (black lines).

Table 5.2: A summary of the uncertainty differences from the A -hyperfine-parameter ratio analysis in Figure 5.4 (second column), and average χ^2_ν values (third column) for the full ^{65,67,69}Ga data sets. Preferential spin assignments are marked with an asterisk (*).

A	σ from ⁷¹ Ga ratio, $I =$			χ^2_ν , $I =$		
	3/2	5/2	7/2	3/2	5/2	7/2
65	1.0*	1.7	2.4	4.6*	5.6	6.8
67	0.0*	0.0*	1.0	7.0*	9.8	12.9
69	3.2*	4.2	10.6	13.6*	28.0	41.2

Shell-model calculations also provide additional evidence for the validity of these assignments. The results using JUN45 and GXPF1 interactions using the KSHELL code [Shi13] can be seen in Figure 5.5. These calculations were performed with an unrestricted model space; GXPF1 calculations provided in this thesis supersedes those in **Article I** which only allowed a maximum of 2p-2h excitations across $N, Z = 28$, due to previous time constraints relating to the publication deadline of the aforementioned article. Even though ⁶¹Ga is currently the lightest odd- A gallium isotope known to be bound with respect to proton emission, ⁵⁹Ga has also been included for completeness of the $N = 28 - 40$ region.

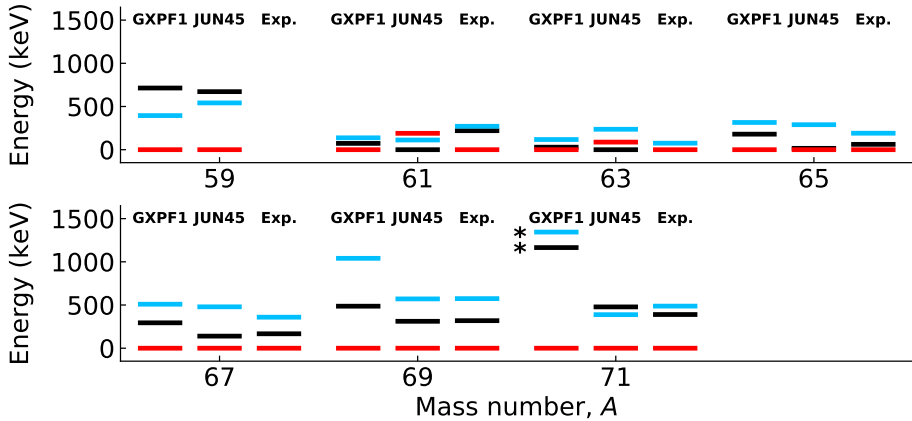


Figure 5.5: GXPF1 (left) and JUN45 (centre) calculations of the lowest-lying $I^\pi = 1/2^-$ (black), $3/2^-$ (red), and $5/2^-$ (blue) states for odd- A isotopes between ^{59–71}Ga inclusive, compared to available literature (right) taken from Ref. [NND18]. States labelled with an asterisk (*) have been offset by 1 MeV.

It is first mentioned that all $I = 1/2$, $3/2$, and $5/2$ levels featuring in the low-lying spectra in Figure 5.5, are predicted, by both the JUN45 and GXPF1 interactions, to originate from a single particle residing in a pf shell-model orbital, which are all associated with a negative parity. Thus for the rest of the spin-assignment discussion regarding $^{65,67,69}\text{Ga}$, nuclear-spin values will be coupled with this type of parity. Overall, both of these interactions predict an $I^\pi = 3/2^-$ ground-state for $^{65,67,69}\text{Ga}$, reinforcing the spin assignments suggested in Chapter 5.3, albeit with almost quasi-degenerate $I^\pi = 1/2^-$ states for JUN45. For $^{61,63}\text{Ga}$ however, this aforementioned interaction ultimately predicts that these $I^\pi = 1/2^-$ nuclear levels manifest as the ground state, contrary to experimental evidence suggesting an $I^\pi = 3/2^-$ ground-state preservation for these isotopes [And05; Pro12]. GXPF1 values correctly predict this observation, however a near-degeneracy of the $I^\pi = 1/2^-$ and $I^\pi = 5/2^-$ nuclear states for odd- A isotopes approaching $N = 28$ suggests mixed-wavefunction configurations. This supports the same conclusion made regarding their g -factor locations in-between the single-particle effective $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ g factors in **Article I**.

5.4 Electromagnetic-moment analyses for $^{65,67,69}\text{Ga}$

Final extracted A - and B -hyperfine parameters for $^{65,67,69}\text{Ga}$ are presented in Tables 5.3 and 5.4. Although these values may differ from those reported in **Article I**, they are consistent with each other and with literature values. Their weighted means have been used for the subsequent calculations of the electromagnetic moments which are graphically displayed in Figure 5.6. The reduction in magnetic-dipole moment uncertainties with respect to those quoted in **Article I** are attributed to using ^{71}Ga reference values from the combined data set in Table 5.1, in contrast to using values from [Che10b] beforehand.

Table 5.3: Extracted A -hyperfine parameters for $^{65,67,69}\text{Ga}$, compared with available literature values. Magnetic-dipole moments have been calculated from a weighted mean of available A -hyperfine parameters; only CRIS-setup values are used for ^{65}Ga . Uncertainties are enclosed within the parentheses.

A	I	$A(4p\ ^2P_{3/2})$ (MHz)		$A(5s\ ^2S_{1/2})$ (MHz)		μ (μ_N)
		CRIS	Ref. [Che10b]	CRIS	Ref. [Che10b]	Combined
65	3/2	+167.5(15)		+939.8(6)		+1.774(1)
67	3/2	+175.3(15)	+175.8(10)	+981.5(17)	+979.7(25)	+1.851(2)
69	3/2	+188.8(8)	+191.5(9)	+1067.5(6)	+1069.5(15)	+2.015(1)

Table 5.4: Extracted B -hyperfine parameters for ^{65,67,69}Ga, compared with available literature values. Electric-quadrupole moments have been calculated from a weighted mean of available B -hyperfine parameters; only CRIS-setup values are used for ⁶⁵Ga. Uncertainties are enclosed within the parentheses.

A	I	$B(4p^2P_{3/2})$ (MHz)		Q (efm ²)
		CRIS	Ref. [Che10b]	Combined
65	3/2	+73(6)		+20.4(17)
67	3/2	+65(4)	+73(4)	+19.4(11)
69	3/2	+62(3)	+63(2)	+17.6(4)

Furthermore, this thesis provides improved GXPF1 calculations for the g factors and electric-quadrupole moments from those reported in **Article I** by using a completely unrestricted model space and allowing a maximum 8p-8h excitations across $N, Z = 28$. Although from Figure 5.6 this amounts to at most a 2.5% change in the g factors for the most neutron-deficient species, the electric-quadrupole moments deviate by almost 25%. These changes are primarily driven by expanding the proton model space because the same action for the neutrons for 4p-4h and above has negligible effects; total convergence for

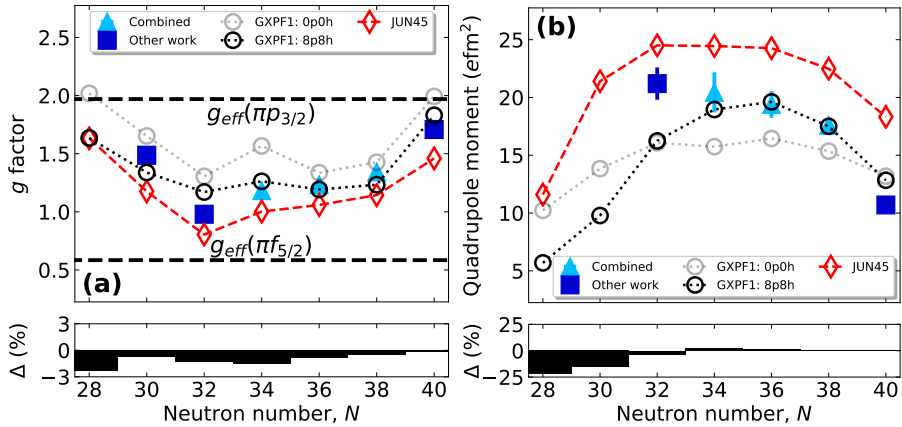


Figure 5.6: **(a)** Extracted g factors, and **(b)** electric-quadrupole moments, for $N = 28 - 40$ gallium isotopes. Combined values from this thesis (light-blue triangles; Tables 5.3 and 5.4) and other literature values (dark-blue squares; Refs. [Wei02; Che10b; Pro12]) are compared to GXPF1 (grey and black circles) and JUN45 (red diamonds) calculations. Differences between 2p-2h (**Article I**) and 8p-8h (these figures) GXPF1 calculations are shown below each figure.

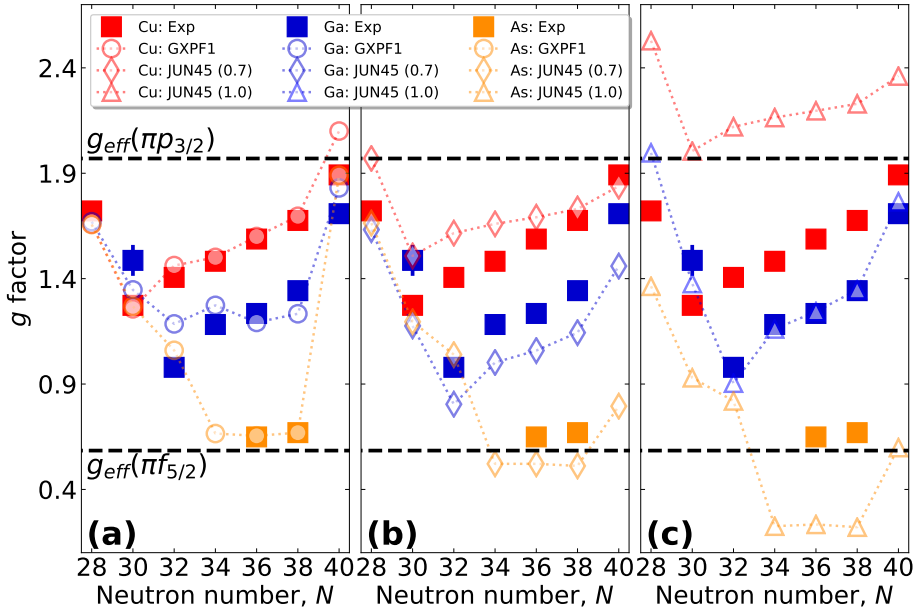


Figure 5.7: Available g factors for the ^{29}Cu (red squares), ^{31}Ga (blue squares), and ^{33}As (orange squares) isotope chains in the $N = 28 - 40$ region, compared with their theoretical counterparts: (a) GXPF1 (circles), (b) JUN45 with $g_s^{eff} = 0.7g_s^{free}$ (diamonds), and (c) JUN45 with $g_s^{eff} = 1.0g_s^{free}$ (triangles).

the electromagnetic moments can already be seen at the 5p-5h level for both proton and neutron model spaces. Their comparison with calculations using a completely restricted model space (0p-0h) also demonstrates the importance of having non-zero $f_{7/2}$ nucleon excitations across both $N, Z = 28$, for replicating the electromagnetic moments using GXPF1 interaction. Overall, Figures 5.6(a) and 5.6(b) show that GXPF1 calculations are better for describing the electromagnetic moments for gallium in this region, reaffirming the indication that ^{56}Ni ($N, Z = 28$) is a soft core.

Exploring the g -factor trends for the neighbouring odd- Z isotope chains in Figure 5.7, those for the ^{29}Cu isotope chain reaffirms the importance of including these types of excitations [Coc09]. Similar conclusions can also be made with the available data for the ^{33}As isotopes. However, it is rather surprising that unquenched JUN45 calculations match almost perfectly with experimental ^{31}Ga g factors with $g_s^{eff} = 1.0g_s^{free}$. The theoretical studies presented in Ref. [Hon04] suggest this to be a unique phenomenon, since the electromagnetic moments for other isotope chains ranging from $Z = 28 - 46$ require the suggested

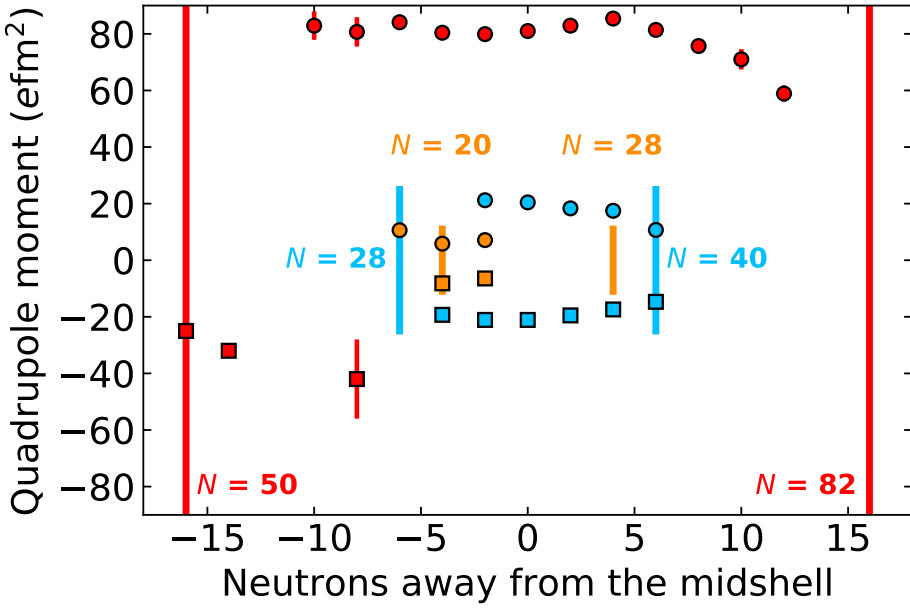


Figure 5.8: Available electric-quadrupole moments for the following proton one-particle/-hole (squares and circles, respectively) isotope chains: ¹⁵Cl [Sun93] and ¹⁹K [Sun93; Min08] with $I^\pi = 5/2^+$ ($\pi 1d_{5/2}$; orange), ²⁹Cu [Vin10; Vin11b] and ³¹Ga [Che10b; Pro12; Far17] with $I^\pi = 3/2^-$ ($\pi 2p_{3/2}$; blue), and ⁴¹Nb [Che09] and ⁴⁹In [Ebe87] with $I^\pi = 9/2^+$ ($\pi 1g_{9/2}$; red). Involved (sub)shell closures for each isotope chain pairs are given by the similarly-coloured and labelled lines, with their mid-points centred at zero.

30% parameter dampening used in Figure 5.7(b). Considering the merits of both JUN45 and GXPF1 calculations for ³¹Ga, further calculations with the A3DA interaction, encompassing the two model spaces, should be performed. Successful electromagnetic moment descriptions for ²⁹Cu [Gro17b] and ³⁰Zn [Wra17] isotopes make it a promising candidate.

Figure 5.8 serves as an extension to Fig. 6 in **Article I** to display the odd-*A* electric-quadrupole moments for other proton one-particle/-hole systems between two (sub)shell closures. Currently, the clear example seen in the ²⁹Cu and ³¹Ga isotope chains between $N = 28$ and $N = 40$ seems to be a unique case; although there are indications between the ⁴¹Nb and ⁴⁹In, and ¹⁵Cl and ¹⁹K isotope chains, additional characterisations are needed where appropriate. Returning to the combined ²⁹Cu and ³¹Ga values in Figure 5.8, the remarkable mirrored trends between odd-*A* isotones (with the same *N*) between $N = 28$ -

40 confirm the linearity expected in (2.23) and (2.24) (see Chapter 2.1.2), as the $\pi 2p_{3/2}$ orbital is filled. As $\pi 2p_{3/2}^1$ and $\pi 2p_{3/2}^{-1}$ systems respectively, they provide confirmation of the validity of shell-model principles with respect to their electric-quadrupole moments. Their parabolic behaviours however, seen as the neutron degree-of-freedom is spanned between the two (sub)shell closures $N = 28$ and 40, are indicative of pn correlation effects that are present and possible core-polarisation effects. Parabolic $B(E2)$ trends have also been observed in the same region with the ^{28}Ni isotope chain [All14]. Other instances can be seen in the ground states of ^{50}Sn isotopes at the mid-point between $N = 50$ and 82 [Eks08], as well as in the isomer states of ^{82}Pb isotopes at the mid-shell point of the $\nu 2g_{9/2}$ orbital [Got12].

5.5 Isotope shifts and changes in the mean-square charge radii for $^{65,67,69}\text{Ga}$

Final isotope-shift values for $^{65,67,69}\text{Ga}$ are presented in Table 5.5. It is reminded from **Article I** that additional problems with the ISOLDE setup prevented the continual point-by-point monitoring of the acceleration voltage and thus, only a single voltage is available for the entire data set: +30011.8 V. Similar difficulties regarding the characterisation of the ISCOOL voltage have been inherent in several COLLAPS analyses [Vin11a; Pro13; Pap15], justifying the need for additional isotope-shift corrections in **Article I** in order to benchmark CRIS-setup values to those reported in previous literature. The inconsistency between the $\delta\nu^{71,69}$ isotope-shift value presented in Table 5.5 with that reported in **Article I**, is attributed to a typographical error in the original analysis procedure. Due to the beleaguered nature of ^{69}Ga hyperfine-structure measurements as explained in Chapter 5.4, its associated isotope shift has

Table 5.5: Isotope shifts and changes in the mean-square charge radii for $^{65,67,69}\text{Ga}$; see text and **Article I** for further details regarding the treatment of systematic problems with the experimental setup. Statistical, modified-systematic, and atomic-systematic uncertainties are enclosed within parentheses, brackets, and braces respectively.

A	I	$\delta\nu^{71,A}$ (MHz)		$\delta\langle r^2 \rangle^{71,A}$ (fm^2)	
		CRIS	Ref. [Pro12]	CRIS	Ref. [Pro12]
65	3/2	+110(5)[1]		-0.412(13)[3]{92}	
67	3/2	+77(4)[1]		-0.252(10)[2]{58}	
69	3/2	+11(2)[1]	+40(4)		-0.116(10){28}

been excluded from re-calibration procedure¹ as detailed in **Article I**; this explains the absence of its change in the mean-square charge radii value in Table 5.5. To within statistical uncertainties, this omission has not affected the new modified isotope shifts for $^{65,67}\text{Ga}$ in Table 5.5, which remain consistent to those published in **Article I**.

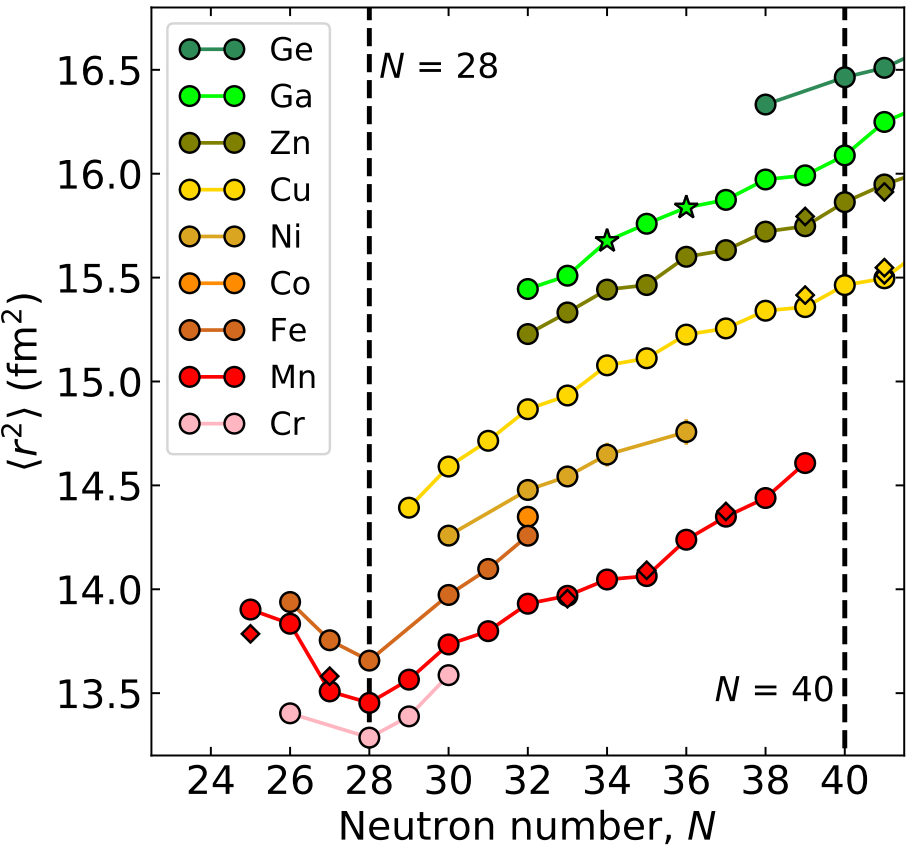


Figure 5.9: Mean-square charge radii for isotopes (ground and long-lived isomer states: circles and diamonds, respectively) in the vicinity of the ^{56}Ni region. Values for $^{65,67}\text{Ga}$ (whose $\delta\langle r^2 \rangle$ have been measured at the CRIS setup; see Table 5.5) are represented with star markers. The $N = 28, 40$ shell/subshell closures are marked with dashed lines. References used for the compilation of this figure can be found in Appendix A.3.

¹Re-calibrated isotope shifts for $^{75,79-82}\text{Ga}$ are presented in Chapter 6.5.

Local behaviour of isotopic mean-square charge radii near ^{56}Ni is displayed in Figure 5.9. These values have been calculated using a reference mean-square charge-radii value from Ref. [Fri04] and the differences for isotope chains currently measured by laser spectroscopy methods. Weighted means have been used where more than one data set is available for a particular isotope chain; values directly from Ref. [Fri04] are used for those chains which have not yet been studied. In particular one striking feature is the sudden jump in mean-square charge radii values for ^{25}Mn and ^{26}Fe at $N = 28$ and is synonymous with expected behaviour when one crosses a shell closure [Cam16]. However, they remain the heaviest isotope chains to assert this characteristic close to ^{56}Ni ; although the gradient steepens in general for the neutron-deficient copper isotopes, additional neutron-deficient measurements from ^{27}Co , ^{28}Ni , ^{29}Cu , ^{30}Zn , and ^{31}Ga are required to further characterise the magic character of ^{56}Ni .

Almost all isotope chains in Figure 5.9 also exhibit OES effects. Generally, any lone nucleon present in the nucleus is responsible by blocking nucleon scattering due to the presence of three- and four-body correlations [Ree71; Zaw85]. For the case of ^{31}Ga , odd- A isotopes appear to have a larger charge radius than their even- A neighbours. Although **Article I** discusses the need for further isotope shift sensitivity to assert this phenomenon with respect to $^{65,67}\text{Ga}$, it is intriguing that an inverted OES occurs at $N = 40$. Recent studies suggest the same manifestation also occurs in the ^{30}Zn isotope chain, albeit as a much weaker effect, and can be explained by the varied proton excitations across $Z = 28$ for the isotopes in this local region [Xie19]. Inversion effects have also been observed as far away as the ^{36}Kr and ^{38}Sr isotope chains below $N = 45$ [Kei95], attributed to strongly deformed states caused by core polarisation from the odd neutron [Lie96]. Further measurements on the ^{32}Ge , ^{33}As , ^{34}Se , and ^{35}Br isotope chains would help to fully characterise the mechanisms involved in OES reversal in the $N = 40$ region with respect to Z evolution. It is remarked that the effects seen in the ^{29}Cu instead originate from a weak subshell effect, which weakens when moving away from $Z = 28$ [Bis16; Xie19]. Similarly, further measurements on the ^{28}Ni isotope chain would help to further characterise this local effect.

5.6 Summary

The measurements of neutron-deficient $^{65,67,69}\text{Ga}$ isotopes have helped to further characterise nuclear structure behaviours near the magic isotope ^{56}Ni . While the g factors for $^{65,67,69}\text{Ga}$ indicate rather mixed configurations, their $I = 3/2$ spin assignments suggested by A -ratio analyses confirm that their configurations are dominated by an odd number of protons in the $\pi 2p_{3/2}$ orbital. However,

these suggestions cannot be completely discerned from the higher-value spin choices. The use of an ionisation scheme providing better sensitivity to spin would help to provide further confidence in assigning spin values.

Electric-quadrupole moment values for odd- A isotopes within $N = 28 - 40$ for ^{29}Cu and ^{31}Ga provide an exceptional demonstration regarding the shell-model expectations for proton one-particle/-hole systems. Observed parabolic trends with the removal of neutron pairs towards $N = 28$ confirm the presence of pn correlations; core-excitation effects may also be prevalent in ^{31}Ga due to a flattened trend at the $N = 34$ midshell. GXPF1 calculations have now been performed for ^{31}Ga isotopes in this region using an unrestricted model space, compared to those published in **Article I**. Although JUN45 calculations reproduce the correct trends, GXPF1 calculations provide a better match to the experimental data overall and provides further evidence of a soft ^{56}Ni core. In particular for the g -factor values, it is surprising that GXPF1 predictions do not perform as well as those for the neighbouring odd- Z ^{29}Cu and ^{33}As isotones. Additional LSSM calculations involving other interactions should be considered in order to fully discern the electromagnetic moment trends for the ^{31}Ga isotopes.

Changes in the mean-square charge radii for $^{65,67}\text{Ga}$ show a continuation of normal OES towards $N = 28$, albeit with a diminished presence. Measurements with greater isotope shift sensitivity would help to confirm this finding, as well as those more neutron-deficient than ^{63}Ga required to confirm a characteristic kink at $N = 28$.

Chapter 6

Nuclear magicity near ^{78}Ni

Conveniently, the next heavier doubly-magic core after ^{56}Ni lies on the same isotopic line: ^{78}Ni . Its positioning at the edge of known observed species in the nuclide chart hitherto made its production at ISOL facilities incredibly difficult and challenging. Studies of local isotopes thus became paramount for indirectly deducing the magicity of ^{78}Ni : spectroscopy of ^{79}Zn [Orl15] and $^{77,79}\text{Cu}$ [Vaj18; Oli17] revealed rather large and robust $N = 50$ and $Z = 28$ shell gaps respectively, for the ^{78}Ni region. Spectroscopy of ^{78}Ni itself has only recently been performed [Tan19], revealing a first-excited 2^+ level at 2600(33) keV and confirming existing theoretical predictions [Hag16]. Plotted alongside neighbouring $E(2_1^+)$ values¹ in Figure 6.1, this firmly cements the status of ^{78}Ni as a magical species.

There is also an abundance of nuclear-structure phenomena in this region of the nuclide chart. Early spectroscopy studies of excited states in the triad $^{69,71,73}\text{Cu}$ revealed a significant lowering of the $I^\pi = 5/2_1^-$ level with respect to the $I^\pi = 3/2^-$ ground state, due to the presence of a monopole term in the residual pn interaction [Fra98; Fra01]. A ground-state $I = 3/2 \rightarrow 5/2$ change later occurs for the ^{75}Cu isotope [Fla09], interpreted as a level-ordering reversal of the $\pi 1f_{5/2}$ and $\pi 2p_{3/2}$ SPE in analogy with theoretical studies on the neighbouring ^{28}Ni isotope chain [Ots10]. While the $I = 5/2$ ground state persists for the next odd- A isotope ^{77}Cu [Kös11; Gro17b] (from in-source and CRIS techniques, respectively), heavier species such as ^{79}Cu have not yet been reached with laser spectroscopy techniques. Coincidentally, this isotope is largely seen as a ‘holy grail’: imagined simply as a $(^{78}\text{Ni} + 1p)$ system, deducing its fundamental properties provides a unique opportunity to indirectly probe ^{78}Ni . Although

¹A similar $E(2_1^+)$ trend in the ^{30}Zn isotopes can also be observed around $N = 50$ [Sha17].

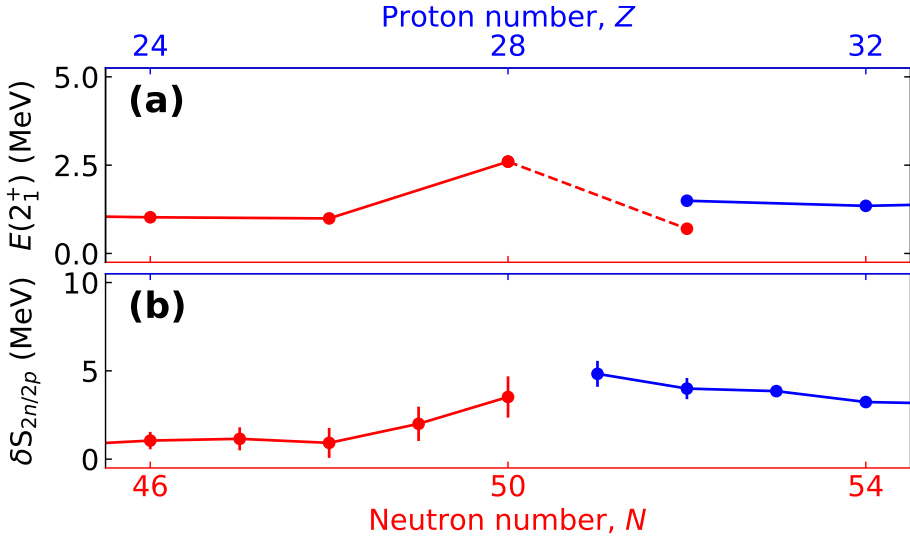


Figure 6.1: **(a)** Energies for the first-excited 2^+ state. **(b)** Two-nucleon-separation energy differences between $N, N+2$ and $Z, Z+2$, for the neutrons and protons respectively. In effect, trends for $Z = 28$ (Ni) isotopes (red) and $N = 50$ isotones (blue) feature in both plots. Data are taken from Ref. [NND18; Tan19]; a theoretical prediction is used for ^{80}Ni (dashed line) [Hag16].

recent decay spectroscopy and mass measurements of this isotope both revealed a significantly large $Z = 28$ shell gap and imply magicity [Oli17; Wel17], nuclear spin and electromagnetic moment information are needed for further clarity. These shell-structure effects are not just confined to ^{29}Cu ; the same ground-state spin change is also observed in the ^{31}Ga isotope chain at $^{81}_{31}\text{Ga}_{50}$ [Che10b], confirming predictions from previous β - and γ -decay spectroscopy works [Hof81; Ver07]. Looking further afield, the study of ^{83}Ga would be considered as highly prized as ^{79}Cu , due to its candidacy² to probe any differences in the expected shell-model $\pi 1f_{5/2}$ orbital description, in the presence of neutron interactions originating from the filling of the the $\nu 2d_{5/2}$ and $\nu 1g_{7/2}$ orbitals, i.e. beyond $N = 50$. First realised with ^{40}Zr isotopes [Sie09], shell-model interactions are also now capable of probing the region beyond $N = 50$. A plethora of studies have benefitted from these recent developments, such as isotopes belonging in the: ^{30}Zn [Als16], ^{31}Ga [Kor13; Kol13; Mad16; Als16; Ver17], ^{32}Ge , [Sie13;

²With a reported ^{79}Cu production rate of 2 ions/ μC using a UC_x target [Wel17] and a "rule-of-thumb" x10 deprecation per mass increment, studying the most obvious candidate ^{81}Cu would currently be practically impossible without further yield or background suppression improvements.

Kor16; Del18], $_{34}\text{Se}$ [Sie13; Cze13; Mat15; Did17; Del18], and $_{36}\text{Kr}$ [Cze13; Did17; Del18] isotope chains for example. With respect to the case for ^{83}Ga , the preservation of the $I^\pi = 5/2^-$ ground state is predicted in analogy with the copper isotopes mentioned above. An experimental confirmation from laser-spectroscopy techniques is thus of utmost importance.

Similar to the structural understanding of ^{79}Cu , the one-neutron-hole nature of $N = 49$ isotones are also good candidates for magicity studies of ^{78}Ni . A low-lying long-lived isomer state has already been identified in ^{80}Ga [Che10a], with a $\nu 1g_{9/2}^{-1}$ structure analogous to those in other $N = 49$ isotones such as in ^{86}Rb [Thi81] and ^{87}Sr [Buc90]. LSSM calculations using the jj44b interaction however, challenge this structural interpretation because only those involving a polarised ^{56}Ni core can successfully and sensibly replicate the isomerism, which would imply a weakened $Z = 28$ shell gap near ^{78}Ni . Subsequent decay-spectroscopy studies [Ver13; Lic14] have aimed to further discern the origin of the two long-lived states in ^{80}Ga by measuring their decay properties, such as their half-lives and branching ratios of transitions that feed into these states. The close proximity between their hyperfine structures [Che10a] however, limited the degree to which their separate decay patterns could be disentangled in Ref. [Ver13], because any laser-ionised beam would most likely have comprised of a mixed-state type. Only studies on isomeric pure-state beams are sufficient for the unambiguous determination of decay properties for each state. Despite the RILIS providing isomer separation for dedicated nuclear-structure studies at ISOLDE [Kös00b; And02; Van04; Ste07; Coc10; Mar13; Pie18; Pie19], this would not be possible for $^{80g,m}\text{Ga}$ because the $\sim \text{GHz}$ linewidth of the RILIS lasers involved [Rot13] would far exceed the isomeric difference as already documented in Ref. [Che10a]. Neither would a Penning-trap mass spectrometer akin to those in mass-measurement experiments [Bol92; Van04; Sta13; Kre13; Alt17; Bab18; Aye19] be appropriate, because the 22.4 keV separation between $^{80g,m}\text{Ga}$ [Lic14] is far below the general³ ~ 100 keV limit [Lun17]. Besides, existing and recent ^{80}Ga mass measurements have failed to resolve this isomer state [Hak08; Aye18; Rei19]; the required $m/\Delta m \geq 3.3 \times 10^6$ mass-resolving power currently remains insurmountable. The high resolution [Gro15] and high selectivity [Lyn16] provided by the CRIS setup however, presents the unique opportunity of individually producing and studying pure-state $^{80g,m}\text{Ga}$ beams.

Furthermore, spectroscopy of these individual beams could provide additional evidence for shape coexistence in the local region. This has already been confirmed with: a long-lived isomer state in ^{79}Zn due to a neutron excitation across $N = 50$ to the $\nu 3s_{1/2}$ orbital [Yan16b], and an intruder 0_2^+ state in ^{80}Ge

³The ISOLTRAP experiment holds the record of measuring the lowest isomeric excitation energy: 33(13) keV with $^{187g,m}\text{Pb}$, achieved with an $m/\Delta m \sim 1.5 \times 10^6$ resolving power [Web05], due to the capability of the RILIS group of producing purified isomeric beams.

which lies lower in energy than the 2_1^+ state [Got16]. Venturing towards the neutron-rich side, the prevalence of long-lived isomerism is completely unknown. Although experimental and theoretical studies provide hints for $^{82,84}\text{Ga}$ [Leb09; Als16], combined laser- and decay-spectroscopy measurements would provide a comprehensive characterisation of any such existences.

6.1 Nuclear-spin-assignment and electromagnetic-moment analyses for $^{75,79,81}\text{Ga}$

In the same manner conducted in Chapter 5.3, Figure 6.2 displays the complete spin analysis for $^{75,79,81}\text{Ga}$ with numerical results in Table 6.1. Likewise, an $I = 1/2$ spin assignment is not considered because more than three resonances are present for each isotope. A cursory look at the the A -hyperfine parameter ratio and χ_ν^2 analyses in Table 6.1 however, reveals difficulties in distinguishing between certain spin assignments, namely ^{79}Ga and ^{81}Ga . Previous gallium studies at the COLLAPS experiment had also encountered similar challenges based solely on the χ_ν^2 analyses; see the Ph.D thesis of E. Mané Jr. in Ref. [Man09a] for further details. A g -factor analysis is thus additionally considered for $^{75,79,81}\text{Ga}$, which compares g factors from A -hyperfine parameters associated with an ambiguous spin assignment, with single-particle estimates for all pf -shell orbitals (using $g_{s,eff} = 0.7g_{s,free}$). This is given in the right-most column of Figure 6.2; individual discussions for each aforementioned isotope now follow.

An $I = 3/2$ spin assignment can be unequivocally assigned for ^{75}Ga from the A -hyperfine-parameter ratio analysis, with a $> 35\sigma$ difference for both $I = 5/2$ and $7/2$. While a g -factor analysis is thus not necessary, it is remarked that its g factor deviates rather significantly from the $\pi 2p_{3/2}$ single-particle estimate; a negative parity is suggested for this state based on its positioning between the $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ single-particle values. Unlike the configuration-mixing scenario in the neighbouring ^{29}Cu isotones with small $M1$ -type ($\pi 1f_{7/2}^{-1} \pi 1f_{5/2}$) $_{1+}$ excitations across $Z = 28$ [Vin11b], these effects are expected to be negligible for the ^{31}Ga isotopes. With an overall greater $\pi 1f_{5/2}$ -orbital occupation, fewer proton excitations from the $\pi 1f_{7/2}$ orbital occur because of Pauli blocking effects [Ney03]. Instead for ^{75}Ga , configurations such as: $[(\pi 2p_{3/2}^2)_{2+} \otimes \pi 1f_{5/2}]_{3/2-}$ with an unaligned proton pair, and $[(\pi 2p_{3/2}^1) \otimes (\pi 1f_{5/2}^2)_{0+}]_{3/2-}$, likely contribute. Its negative electric-quadrupole moment (see Chapter 6.2) further supports the presence of the latter contribution, suggesting a $\pi 2p_{3/2}$ single particle mixed with a $\pi 1f_{5/2}$ structure⁴, akin to ^{29}Cu isotopes between $N = 28$ and $N = 40$.

⁴The relation: $Q_s(A_{29}\text{Cu}_N) = -Q_s(A_{31}^{+2}\text{Ga}_N)$, is reminded from Figure 5.8.

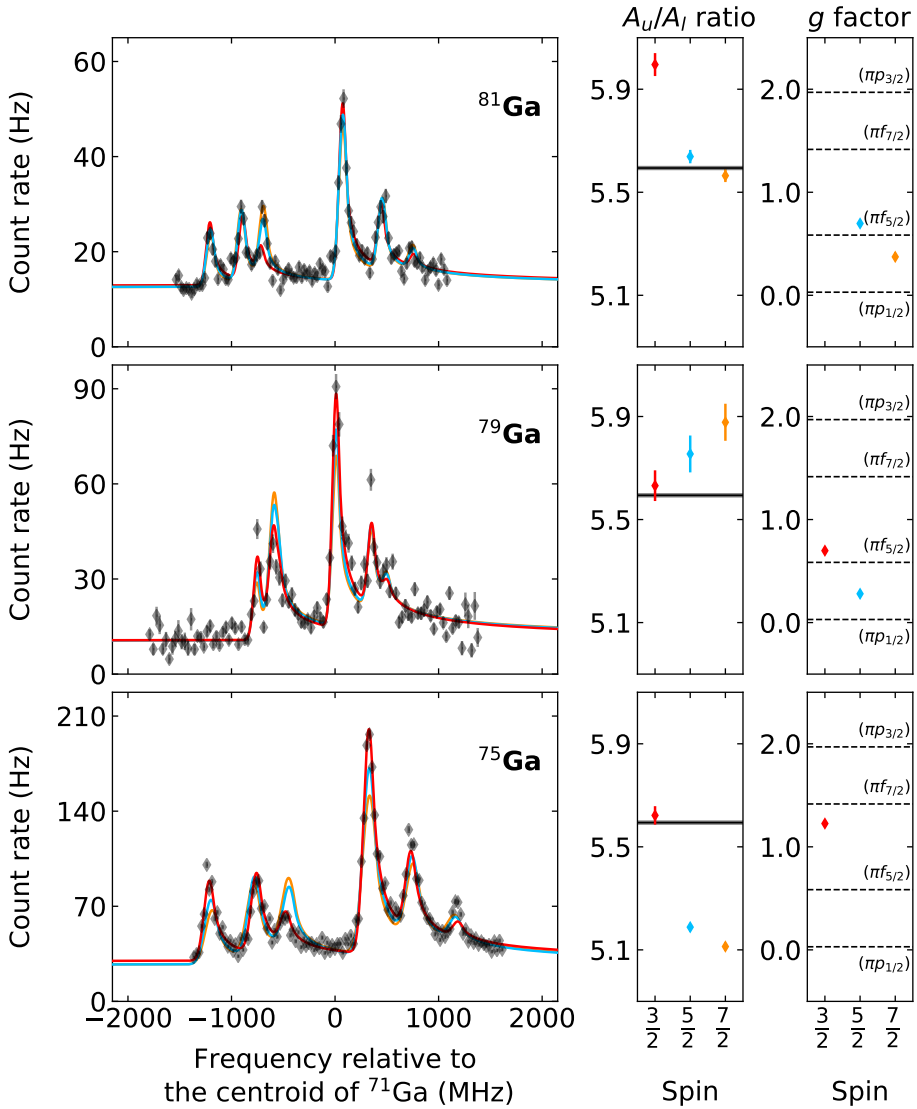


Figure 6.2: Selected hyperfine spectra of $^{75,79,81}\text{Ga}$, each fitted assuming $I = 3/2$ (red), $5/2$ (blue), and $7/2$ (orange) spin assignments. Corresponding A -hyperfine-parameter ratios and g factors (where appropriate) extracted from the complete data set for each spin assignment are displayed in the middle and right column, compared with the CRIS-setup ^{71}Ga A -hyperfine-parameter ratio from Table 5.1 (black lines) and effective single-particle g factors (dashed lines), respectively.

Table 6.1: A summary of the uncertainty differences from the A -hyperfine-parameter ratio analysis in Figure 6.2 (second column), and average χ^2_ν values (third column) for the full $^{75,79,81}\text{Ga}$ data sets. Preferential spin assignments are marked with an asterisk (*).

A	σ from ^{71}Ga ratio, $I =$			χ^2_ν , $I =$		
	3/2	5/2	7/2	3/2	5/2	7/2
75	0.7*	37.4	36.4	18.0*	37.4	56.3
79	0.6*	2.0	3.8	4.6*	5.7	6.8
81	8.1	1.5	1.1*	3.6	2.7*	3.1

Likewise, an A -hyperfine parameter analysis for ^{79}Ga also suggests an $I = 3/2$ spin assignment, however $I = 5/2$ cannot be definitively excluded with a $\sim 2\sigma$ distance. This isotope highlights the necessity of a g -factor analysis because it is able to show that only the $I = 3/2$ g factor corresponds to a single-particle value, albeit belonging to the $\pi 1f_{5/2}$ orbital. It can be interpreted as a seniority-3 ($\pi 1f_{5/2}^3$) $_{3/2^-}$ configuration [Che10b], thus also suggesting a negative-parity state; such configurations have also been reported in the neighbouring ^{30}Zn isotope chain [Wra17; Yan18b] and is indicative of deviations from expected single-particle behaviour. An unexpected change in its sign and value (see Chapter 6.2), with respect to that for $^{75,77}\text{Ga}$ beforehand, also confirms the continuation of a changing shell structure.

In contrast, ^{81}Ga is expected to exhibit single-particle proton behaviour because its magic number ($N = 50$) of neutrons should not contribute towards the nuclear-structure properties. Although an 8.1σ deviation in the A -hyperfine parameter ratio for $I = 3/2$ confirms a spin-value change at $N = 50$, there is minimal distinction between $I = 5/2$ and $7/2$. Only the g -factor analysis can confirm an $I = 5/2$ assignment because its corresponding g factor agrees very favourably with the single-particle estimate for the $\pi 1f_{5/2}$ orbital, unlike that for $I = 7/2$ compared with $\pi 1f_{7/2}$. Akin to the cases for $^{75,79}\text{Ga}$, this also suggests a negative ground-state parity. Additional (shell-model) evidence against a ground-state $I = 7/2$ spin assignment is also provided by the energetically unfavourable 2p-1h excitation across $Z = 28$ that would otherwise be needed. Its small electric-quadrupole moment ($Q_s = -4.6(2)$ efm²) in Table 6.3 and Figure 6.3(b) (see Chapter 6.2) is also expected for an isotope at a shell closure, in which non-deformed spherical shapes are predicted by the shell model.

Overall, this combined analysis has assigned $I^\pi = 3/2^-, 3/2^-,$ and $5/2^-$ spin and parity values for the ground states of $^{75,79,81}\text{Ga}$ respectively; this is consistent with the findings reported in Ref. [Che10b]. It is remarked that both studies necessitated additional analysis in order to make definitive spin assignments

for: ⁷⁹Ga for the COLLAPS experiment in Ref. [Che10b], and ⁸¹Ga for the CRIS experiment in Ref. [Far17] and in this thesis. Altogether, both these studies complement each other by providing an enhanced certainty for the spin assignments of these isotopes. Despite the additional g -factor analysis providing a useful tool for inferring the spin assignments for neutron-rich odd- A gallium isotopes in this thesis, it is also reminded that this had been necessary due to the indistinguishability of some of the A -hyperfine parameter ratio analyses for certain isotopes. Especially if heavier neutron-rich species were to be measured in the future, the prestige of laser-spectroscopy techniques could potentially be undermined with a shell-model dependency. One possible solution would be to use a transition scheme which is more sensitive to the nuclear spin; this is explored further in Chapter 8.1.3.

6.2 A summary of the hyperfine parameters and electromagnetic moments for ^{75,79,81}Ga

Based on the spin assignments made in Chapter 6.1, final extracted hyperfine parameters for ^{75,79,81}Ga are presented in Tables 6.2 and 6.3; magnetic-dipole and electric-quadrupole moment calculations have been extracted from the $A(4p\ ^2P_{3/2})$ and $B(4p\ ^2P_{3/2})$ parameters respectively. These moments are also graphically displayed Figure 6.3. While JUN45 calculations can still be made for these isotopes, this is not option for GXPF1 due to the non-inclusion of the $0g_{9/2}$ orbital in its model space. Instead, previous studies have compared with values calculated from the jj44b interaction [Che10b] and those are also included in Figure 6.3. Although calculations for the neutron-deficient isotopes can be done with this interaction, they do not feature in Chapter 5.4 because of its limited accessibility.

Table 6.2: Extracted A -hyperfine parameters for ^{75,79,81}Ga, compared with available literature values. Magnetic-dipole moments have been calculated from a weighted mean of available A -hyperfine parameters. Uncertainties are enclosed within parentheses.

A	I	$A(4p\ ^2P_{3/2})$ (MHz)		$A(5s\ ^2S_{1/2})$ (MHz)		μ (μ_N)
		CRIS	Ref. [Che10b]	CRIS	Ref. [Che10b]	Combined
75	3/2	+172.4(11)	+173.6(9)	+972.3(7)	+973.1(15)	+1.835(1)
79	3/2	+98.6(16)	+98.3(9)	+553.5(40)	+555.0(14)	+1.047(1)
81	5/2	+98.4(3)	+98.9(4)	+554.5(9)	+555.6(13)	+1.745(2)

Table 6.3: Extracted B -hyperfine parameters for $^{75,79,81}\text{Ga}$, compared with available literature values. Electric-quadrupole moments have been calculated from a weighted mean of available B -hyperfine parameters. Uncertainties are enclosed within the parentheses.

A	I	$B(4p^2P_{3/2})$ (MHz)		Q (efm ²)
		CRIS	Ref. [Che10b]	Combined
75	3/2	-109(1)	-105(1)	-30.1(9)
79	3/2	+49(4)	+58(2)	+15.9(10)
81	5/2	-16(2)	-18(3)	-4.6(2)

A comparison of the odd- A gallium g factors between $N = 40 - 50$ with the shell-model calculations in Figure 6.3(a) shows generally good agreements with all three JUN45, jj44b, and fpg interactions. Considering the absences of: proton excitations across $N = 28$ from the $\pi 1f_{7/2}$ orbital for the JUN45 and jj44b interactions, and nucleon excitations to the $1g_{9/2}$ orbitals for the fpg interaction, this could suggest these are not required for the description of their g factors. It is noted that the deviation for the g factor of ^{73}Ga is due to the collective nature of its $I = 1/2$ ground state, with a very mixed wave-function

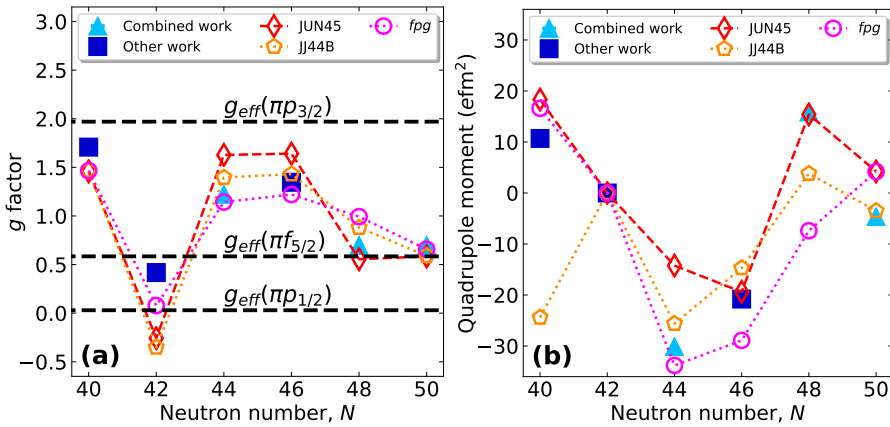


Figure 6.3: (a) Extracted g factors and (b) electric-quadrupole moments for odd- A gallium isotopes from $N = 40 - 50$ inclusive. Values presented in this thesis (light-blue triangles) and other literature values (dark-blue squares; Refs. [Che10b]) are compared to JUN45 (red diamonds), jj44b (orange pentagons; Ref. [Che10b]) and fpg (pink circles; Ref. [Sri11]) calculations.

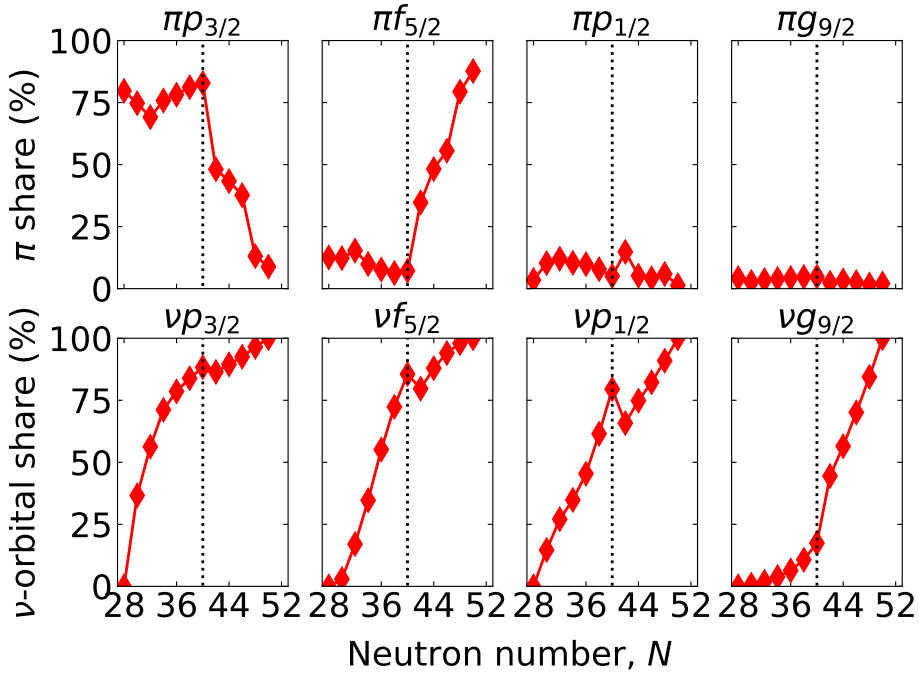


Figure 6.4: (Top row) The distribution of the three valence protons (${}_{31}\text{Ga}$: $Z = 28 + 3p$), and (bottom row) the occupancy of the neutron orbitals, across the orbitals involved in the JUN45 model space for the ground states of odd- A ${}^{59-81}\text{Ga}$ inclusive. The $N = 40$ sub-shell closure is marked with a dotted black line in each subplot.

due to the closeness of the $\pi 1f_{5/2}$ and $\pi 2p_{1/2}$ single-particle levels [Hon09]; only the fpg interaction predicts the correct sign. Looking at the JUN45-predicted proton and neutron occupancies from within the pf shell in Figure 6.4, a sudden proton depletion and filling in the $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ orbitals respectively after $N = 40$ also supports the beginnings of a changing shell-structure at $N = 42$ as described in Chapter 6.1. A further sudden jump at $N = 48$ corresponds with a structural transformation starting in ${}^{79}\text{Ga}^5$, despite the $I = 3/2 \rightarrow 5/2$ switch occurring at $N = 50$ for ${}^{81}\text{Ga}$ in Chapter 6.1, with its somewhat collapsed hyperfine structure in Figure 6.2 indicative of this. With 79% and 88% of the valence protons residing in the $\pi 1f_{5/2}$ orbital for ${}^{79}\text{Ga}$ and ${}^{81}\text{Ga}$ respectively, shell-model calculations also show their nuclear structure should be dominated by a $\pi 1f_{5/2}^3$ configuration.

⁵For both JUN45 and jj44b interactions, it is the first-excited $I^\pi = 3/2^-$ state in ${}^{79}\text{Ga}$ that matches the experimental electromagnetic moments.

For the electric-quadrupole-moment comparisons in Figure 6.3(b), a greater variation between the aforementioned shell-model calculations can be observed. Overall, the fp g interaction performs much better for the isotopes toward the $N = 40$ subshell closure (*id est* $^{71,73,75}\text{Ga}$); calculations for the $N \leq 40$ gallium isotopes are strongly recommended for any associated future laser-spectroscopy studies, to see if they match better than those borne from the GXPF1 interaction. Incorrect signs are predicted for $^{79,81}\text{Ga}$ residing close to $N = 50$ however, and could be indicative of missing proton excitations across $Z = 28$ from the $\pi 1f_{7/2}$ orbital [Sri11], which would be reflected in the electric-quadrupole moments. JUN45 calculations are a better match for the neutron-rich species ($^{77,79,81}\text{Ga}$), where some of the most dramatic nuclear structure changes for the gallium isotope chain take place, as previously discussed in the section above. Calculations using the jj44b interaction can be considered comparable to JUN45 in this region; it is the only interaction which predicts the correct sign for the electric-quadrupole moment of ^{81}Ga . The use of a shell-model interaction that encapsulates the entirety of all three model spaces for the calculations used in this section is desired in order to further probe the more intricate nuclear structure details displayed by the electric-quadrupole moments.

6.3 Nuclear-spin-assignment and electromagnetic-moment analyses for $^{80,82}\text{Ga}$

Measurements of odd-odd nuclei are increasingly seen as more important for the characterisation of doubly-magic nuclei, compared to their odd- A counterparts. Despite an inherent increase in difficulty with respect to their calculations and interpretations, they can be used to probe the effects of pn correlations near doubly-magic systems; see studies on ^{78}Cu for probing ^{78}Ni [Gro17a]. With respect to the $_{31}\text{Ga}$ isotope chain, $^{80,82}\text{Ga}$ represent two such odd-odd probes; however unlike the well-defined structures of odd- A gallium isotopes presented in the prior sections, numerous even- A variants are known to possess collapsed hyperfine structures for the 417.2-nm transition [Man11a]. This is also true for $^{80,82}\text{Ga}$; not only apparent in Refs. [Che10a; Che12a] but also demonstrated in the upcoming figures (see Figures 6.7 and 6.11). Despite these complexities, a combination of shell-model techniques and new data in the neighbouring isotopes can be used to bring further insight into their structures.

6.3.1 Paar's parabola rule predictions

Before looking at the analysis of CRIS-setup hyperfine-spectroscopy measurements for $^{80,82}\text{Ga}$, Paar parabola predictions are first examined in order to discern the likeliest spin for their ground states. It is stated that the purpose of this exercise (which is also applicable for the ^{214}Fr case study presented later in Chapter 7.2.1) serves to only provide an indication for the spin assignments, which does not fully take into account any additional correlation and mixing effects.

Figure 6.5 shows Paar parabolas for suspected pn configurations in ^{80}Ga based on the likeliest shell-model orbitals to be involved from Figure 2.1 and from the analysis in Chapter 6.1: $\pi 1f_{5/2}$ ($\pi 1f_{5/2}^3$; half-filled orbital), $\pi 2p_{3/2}$ ($\pi 2p_{3/2}^3$; hole configuration), and $\nu 1g_{9/2}$ ($\nu 1g_{9/2}^9$; hole configuration). It is assumed that the $\pi 1f_{5/2}$ shell is more bound with respect to $\pi 2p_{3/2}$ based on the conclusions made in Chapter 6.1, as well as its proton-occupancy domination as documented in Figure 6.4 and in Ref. [Man11a]. As a first-order estimation,

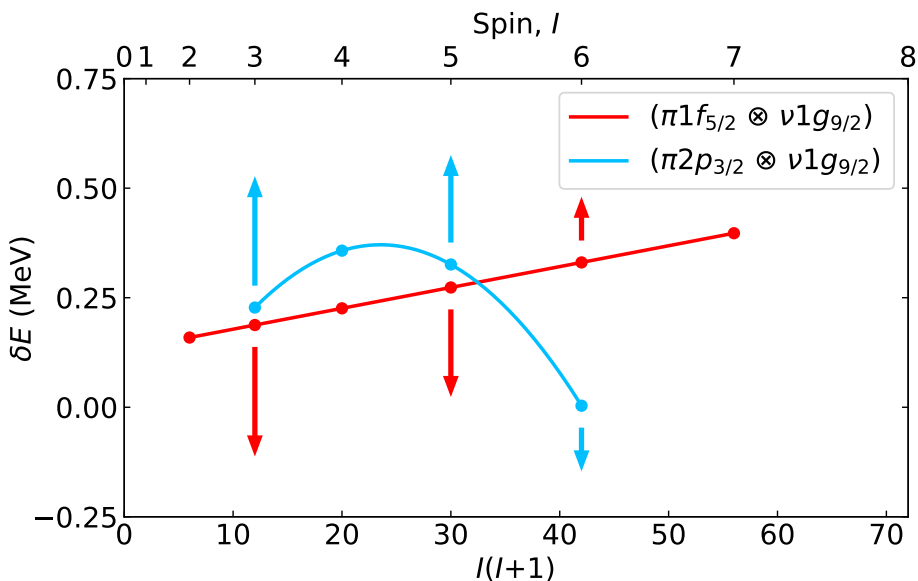


Figure 6.5: Paar parabolas for the nuclear structure of ^{80}Ga , assuming: $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ (red), and $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ (blue) configurations. The $\pi 2p_{3/2}$ SPE is shifted with respect to that for $\pi 1f_{5/2}$ by 350.6 keV. The arrows visualise instances of how the mixing between alike spin states can repel certain states to lower energies; see text for further details.

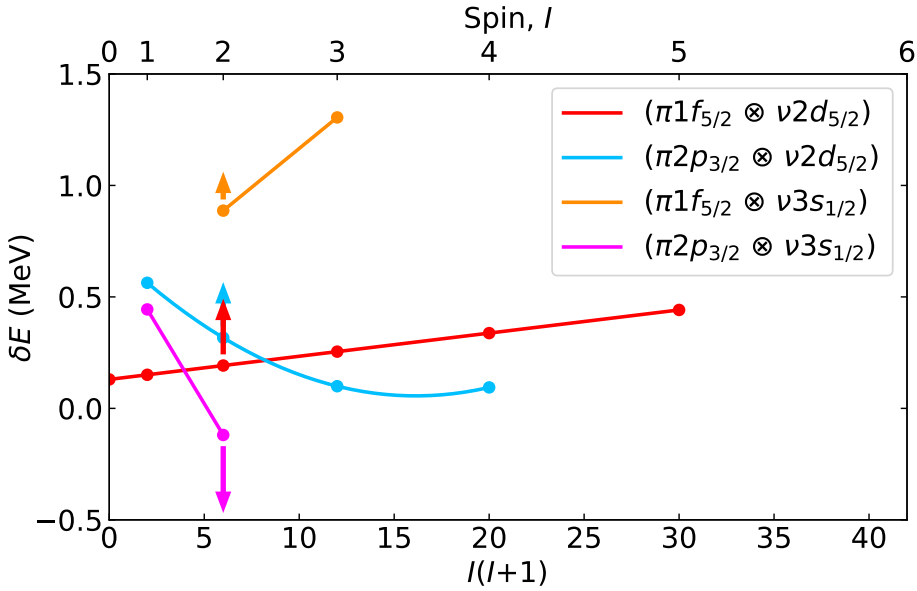


Figure 6.6: Paar parabolas for the nuclear structure of ^{82}Ga , assuming: $(\pi 1f_{5/2} \otimes \nu 2d_{5/2})$ (red), $(\pi 2p_{3/2} \otimes \nu 2d_{5/2})$ (blue), $(\pi 1f_{5/2} \otimes \nu 3s_{1/2})$ (orange), and $(\pi 2p_{3/2} \otimes \nu 3s_{1/2})$ (magenta) configurations. The $\pi 2p_{3/2}$ SPE is shifted with respect to that for $\pi 1f_{5/2}$ by 350.6 keV, while the $\nu 3s_{1/2}$ SPE is shifted with respect to that for $\pi 2d_{5/2}$ by 388.0 keV. The arrows visualise instances of how the mixing between alike spin states can repel certain states to lower energies; see text for further details.

a 350.6 keV offset has been applied to the $\pi 2p_{3/2}$ orbital, based on recent ^{81}Ga (with $I_{gs}^{\pi} = 5/2^{-}$) γ -decay spectroscopy results in which the first-excited $I^{\pi} = 3/2^{-}$ state, predominantly arising from a single proton excitation, has been observed at that energy [Dud19]. Additionally, all states described in this section henceforth are associated with negative parity, due to their association with the pn configurations considered for ^{80}Ga . It is also remarked that Paar-parabola analyses have also been performed for other $N = 49$ isotones such as ^{82}As , ^{84}Br , and ^{86}Rb , see Ref. [Eti15]. The overall analysis predicts an $I^{\pi} = 6^{-}$ state for the ground state, arising from the $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ configuration. However additionally, the $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ and $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ configurations are quasi-degenerate for the $I^{\pi} = 3^{-}$ and 5^{-} states. States from these configurations are thus likely to mix and repel one another, whereby the levels associated with the latter configuration are pushed towards lower energies. The competition between the $I^{\pi} = 3^{-}$, 5^{-} , and 6^{-} states means that they are likely to feature as low-lying levels in ^{80}Ga .

The analogous situation for ^{82}Ga is shown in Figure 6.6, based on the following shell-model orbitals: $\pi 1f_{5/2}$ ($\pi 1f_{5/2}^3$; half-filled orbital), $\pi 2p_{3/2}$ ($\pi 2p_{3/2}^3$; hole configuration), $\nu 2d_{5/2}$ ($\nu 2d_{5/2}^1$; particle configuration), and $\nu 3s_{1/2}$ ($\nu 3s_{1/2}^1$; half-filled configuration). Assuming the same proton-shell ordering for the case in ^{80}Ga in Figure 6.5, the picture for the neutron orbits considers a more bound $\nu 2d_{5/2}$ shell with respect to $\nu 3s_{1/2}$, based on the systematics for ^{81}Zn and ^{83}Ge [Ver07; Als16]. A 248.2 keV offset has thus been applied to the $\pi 3s_{1/2}$ orbital, based on recent ^{83}Ge (with $I_{gs}^\pi = 5/2^+$) γ -decay spectroscopy results in which the first-excited $I^\pi = 1/2^+$ state is thought to arise from a single neutron excitation, from the latter aforementioned reference. While initially the picture may look rather complex, it is remarked that an $I^\pi = 2^-$ state can be created from all the pn configurations considered in Figure 6.6. In particular, the majority of these states are somewhat clustered together, meaning they are likely to all mix and strongly repel with each other. Thus, this analysis predicts an $I^\pi = 2^-$ ground state for ^{82}Ga , albeit with a very mixed configuration.

6.3.2 Two long-lived states in ^{80}Ga

The first focus of ^{80}Ga is the central collapsed structure which is believed to be the ground-state; comparisons between extracted electromagnetic moments and those predicted by JUN45 and jj44b calculations infer an $I^\pi = 6^-$ assignment [Che10a]. In an effort to ultimately be able to employ shell-model-independent techniques, one measurement within the arsenal of ^{80}Ga CRIS-setup scans (from the 2015 experiment) focused solely on this ground-state structure and used 417.2-nm light with a reduced power density to reduce the peak linewidths. A cursory look at the resulting sub-structure presence in Figure 6.7 with a FWHM of 76(4) MHz rules out an $I = 0$ assignment. However despite this achievement, extracted A -hyperfine parameter ratios for suspected spin assignments are neither distinguishable from one another, nor consistent with the accepted ^{71}Ga A -ratio value of +5.594(5) in Chapter 5.2.

Akin to the methodology of Chapter 6.1, g -factor analyses are performed instead, extracted from different spin-assignment fits with constrained A -ratios and intensities as predicted from (2.45). They have been compared to empirical values calculated assuming pure ($\pi 1f_{5/2} \otimes \nu 1g_{9/2}$) and ($\pi 2p_{3/2} \otimes \nu 1g_{9/2}$) configurations with (2.17). Proton and neutron single-particle g factors are taken from magnetic-dipole moment values for nearby $^{79,81}\text{Ga}$ (this thesis; Table 6.3) and ^{79}Zn [Yan16b] respectively, the latter of which has only recently been made available. Figure 6.7 shows the results of this analysis, for the possible spin-value range of $I = 2 - 7$. From a brief examination of the hyperfine-spectroscopy fits, an $I = 2$ assignment can probably be ruled out because of its stark difference

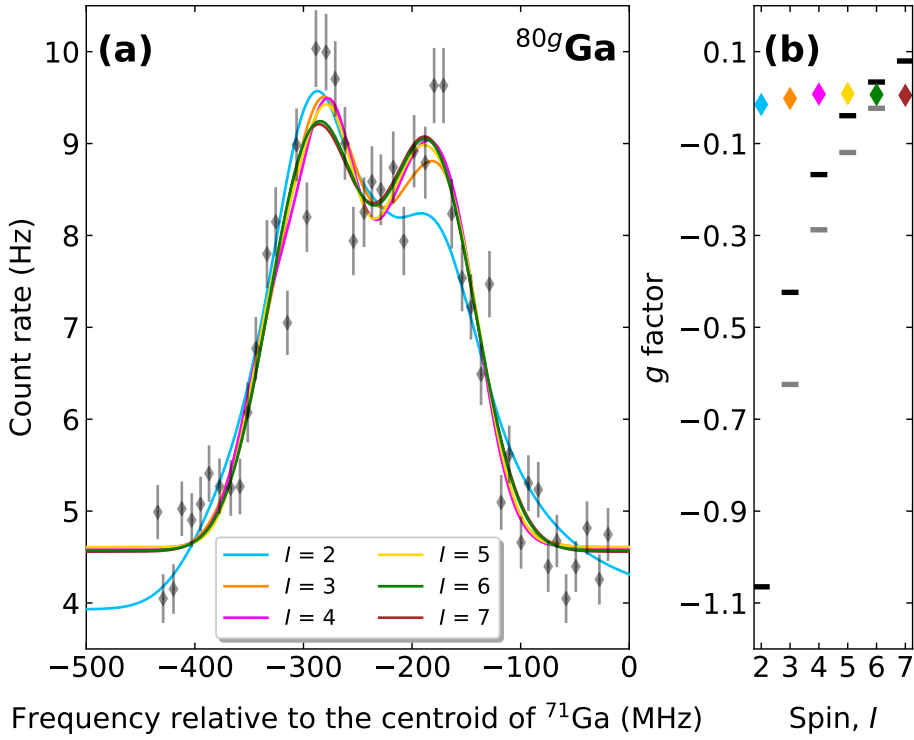


Figure 6.7: **(a)** A hyperfine spectrum of ^{80g}Ga , fitted assuming $I = 2$ (blue), 3 (orange), 4 (magenta), 5 (yellow), 6 (green), and 7 (brown) spin assignments. **(b)** Extracted g factors compared with applicable empirical values assuming pure: $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ (black lines), and $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ (grey lines) configurations.

with respect to the other presented fits, in particular regarding the predicted background level. From the g -factor analysis, the strongest preferences can be seen with $I = 5, 6$, and 7 assignments; all other choices lie $> 25\sigma$ away from their respective empirical calculations and can be unequivocally ruled out from this exercise. It is also remarked that the g factor for $I = 6$ is the only value that can be described by both pn configurations mentioned in Figure 6.7: a 52%:48% composition of the $\pi 1f_{5/2}$ and $\pi 2p_{3/2}$ configurations respectively.

Extending the frequency space, an isomeric state of ^{80}Ga can also be observed surrounding the structure of ^{80g}Ga and has been a contentious topic of debate regarding its existence [Hak08; Che10a; Lic14; Ver13]. Likewise with ^{80g}Ga , the observation of more than one peak definitively rules out $I = 0$; additionally,

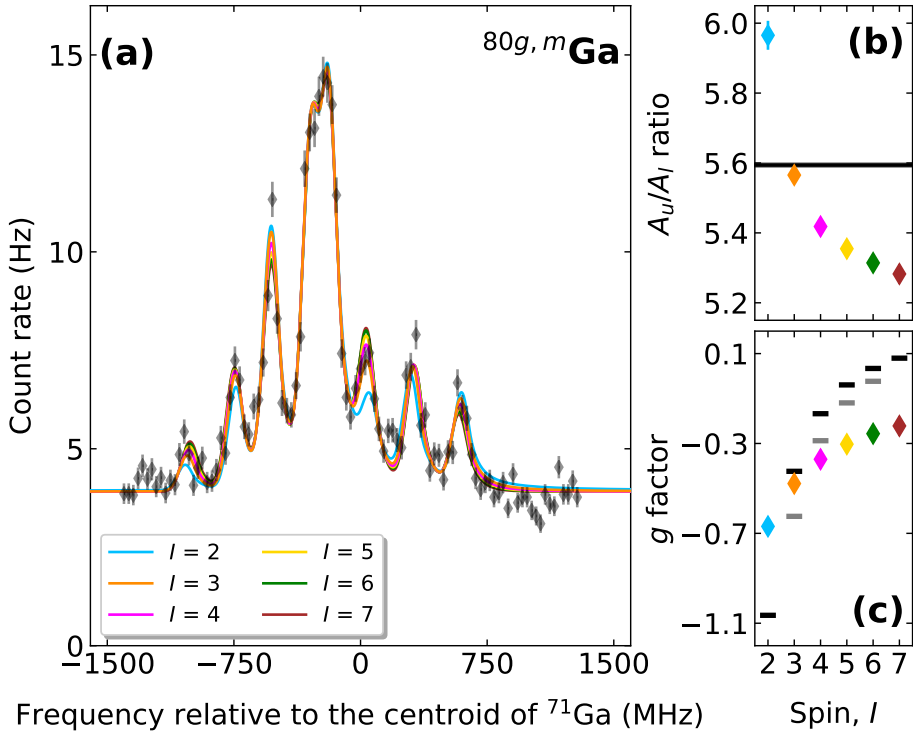


Figure 6.8: (a) A hyperfine spectrum of $^{80g,m}\text{Ga}$ (central and wider structures) fitted assuming $I = 2$ (blue), 3 (orange), 4 (magenta), 5 (yellow), 6 (green), and 7 (brown) spin assignments. (b) Extracted A -hyperfine-parameter ratios for ^{80m}Ga compared with that for ^{71}Ga from Table 5.1 (black line). (c) Extracted ^{80m}Ga g factors compared with applicable empirical values assuming pure: $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ (black lines), and $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ (grey lines) configurations.

a total observation of six resonances also excludes an $I = 1$ spin assignment. However its wider structure allows for an A -hyperfine parameter ratio analysis to be performed like in Chapters 5.4 and 6.2. From Figure 6.8, an $I = 3$ assignment is strongly preferred with all other spin-value options at least 6σ away from the accepted ratio. This predilection also arises with the g -factor comparison in which it is the only value that can be described by the two $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ and $(\pi 2p_{3/2} \otimes \nu 1g_{9/2})$ configuration, with a 73%:27% mix required. It is remarked that the differing intensities between the ground-state structures observed in Figures 6.7 and 6.8 are attributed to power-broadening effects due to the different laser powers used for these scans. This explains why

either χ^2_ν or hyperfine-intensity comparisons would not be appropriate for any spin-value assignment of these isotopes.

The additional consideration that several LSSM calculations already predict the manifestation of a low-lying $(3,4,5,6)^-$ multiplet [Che10a] also favours the $I^\pi = 3^-$ and 6^- suggestions made in the analysis above, both with a negative parity. It is noted that $I^\pi = 5^-$ could also be considered as a ground-state candidate from Figure 6.7, as well as from the Paar parabola analysis in Figure 6.5. However this assignment has already been discounted in Ref. [Che10a] because otherwise, the resulting $E2$ transition between $3^- \rightarrow 5^-$ states would make the former state too short lived to be isomeric. Therefore, this further strengthens the case for an $I^\pi = 6^-$ ground-state as alluded from the analysis in Figure 6.7. Overall, the $I^\pi = 6^-$ and 3^- ground- and isomer-state spins for the two low-lying levels in ^{80}Ga agree with the Paar parabola analysis presented in Chapter 6.3.1, confirming the usefulness of its methodology. Differing interpretations regarding the expected characters for each state (a mixed $I^\pi = 3^-$ state and a pure $I^\pi = 6^-$ state with Paar; *vice versa* in the g -factor analyses in Figures 6.7 and 6.8) however, reflects the approximate nature of the Paar-parabola analysis as stated in the aforementioned chapter. Despite this convincing experimental nuclear-structure evidence nonetheless, laser-spectroscopy techniques cannot provide any further insights regarding its nuclear structure in addition to those already provided in Ref. [Che10a]. More robust conclusions could be made by studying a transition with a greater hyperfine-state splitting and performing decay-spectroscopy studies on each state individually; these will be explored further in Chapters 8.1.3 and 8.1.4.

6.3.3 The nuclear structure of ^{82}Ga

^{82}Ga currently remains the heaviest gallium isotope to have been analysed with laser-spectroscopy methods. Based on a strong β -decay feeding to the 2^+ levels in ^{82}Ge , previous studies have suggested a low-value ground-state spin in the range $I = (1,2,3)$ [Hof81]. Recent laser spectroscopy studies tentatively assigned it $I = (2)$ [Che12a]; however likewise with ^{80g}Ga , it exists as a collapsed structure.

Total summed data using two different ISCOOL accumulation times can be seen in Figure 6.9. Intriguingly for the summed scans collected using a longer bunch period, the central and right-most peaks look to be slightly suppressed in intensity which could imply the presence of a shorter-lived isomeric state residing in the structure. Although Refs. [Kam12; Als16] allude to the existence of isomeric states in ^{82}Ga , their corresponding $< \mu\text{s}$ half-lives would not allow them to be detected by laser spectroscopy techniques; thus any indication of

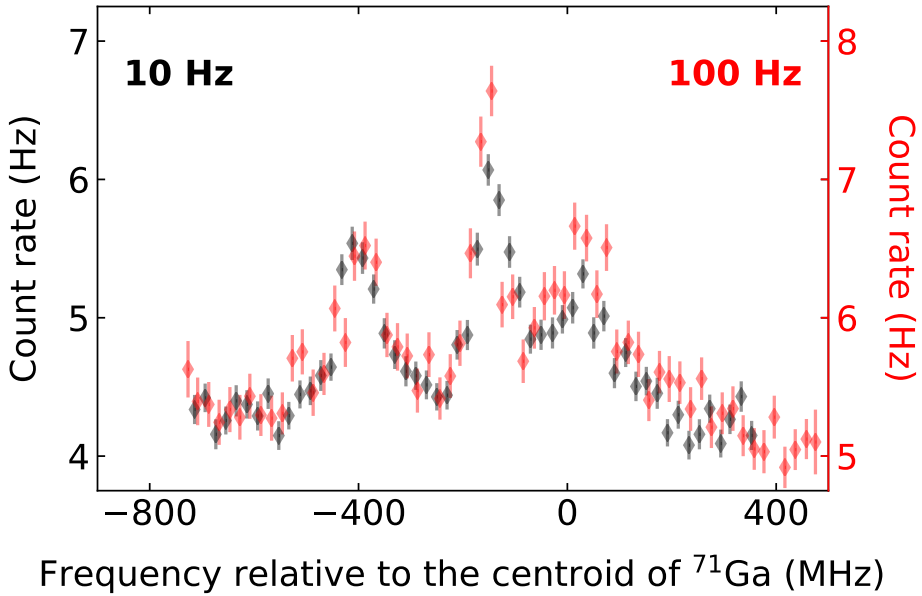


Figure 6.9: Hyperfine spectra of ^{82}Ga using ISCOOL collection times of 100 ms (black; 10 Hz) and 10 ms (red; 100Hz). Both are normalised with respect to their background count rates deduced from fits assuming $I = 2$ spin assignments.

isomeric states in this study would be unexplained in these previous decay spectroscopy works. To investigate this further, χ^2 values were obtained from fits using different A_u and B_l values for each spin assignment, while fixing the A -hyperfine parameter ratio to the value quoted in Table 5.1 and minimising all other fit parameters. For a particular spin assignment, the standard deviation of any (A_u, B_l) pair away from the optimal set can be determined by the square root of their χ^2 difference [Gin18]; these plots are shown in Figure 6.10. For each spin assignment from $I = 1$ to 5, regions of low-value standard deviations appear to manifest in the same regions between the two accumulation times and does not indicate any anomalous behaviour. The hindrance in this discussion can be attributed to the large uncertainties seen in Figure 6.9, despite nearly 9 hours of collected data for each ISCOOL setting. Thus at present, there is no clear evidence of isomeric structure in ^{82}Ga and means that its full analysis in Figure 6.11 combines both data sets featuring in Figure 6.9.

Mirroring the analysis performed for $^{80g,m}\text{Ga}$ in Chapter 6.3.2, the g -factor analysis assumes that both $\pi 1f_{5/2}$ and $\pi 2p_{3/2}$ orbitals can contribute. Although the most obvious choice for the lone valence neutron past $N = 50$ is to reside

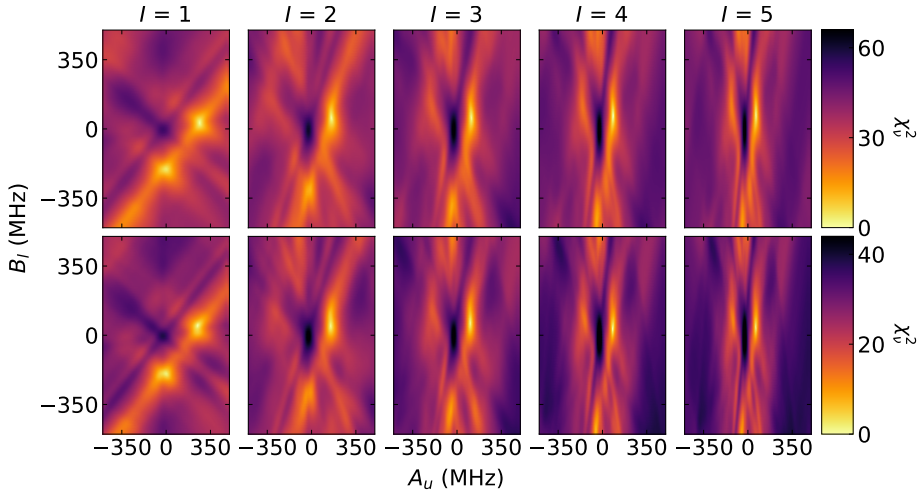


Figure 6.10: Goodness-of-fit plots resulting from fitting the observed ^{82}Ga hyperfine spectra in Figure 6.9 (top row: 10 Hz, bottom row: 100 Hz), with respect to fixed A_u - and B_l -hyperfine parameters. See text for further details.

in the $\nu 2d_{5/2}$ orbital, β -decay studies on ^{83}Ge have confirmed a narrowing of the energy gap between the $\nu 2d_{5/2}$ and $\nu 3s_{1/2}$ orbitals for the $N = 51$ isotones as $Z = 28$ is approached [Per06]. Thus, both neutron orbitals are considered: altogether, four pn contributions can be made, which are stated in Figure 6.11. While single-particle g factors for the $\pi 2p_{3/2}$, $\pi 1f_{5/2}$ and $\nu 3s_{1/2}$ orbitals are readily available from nearby $^{79,81}\text{Ga}$ (this thesis; Table 6.3) and ^{79m}Zn [Yan16b] respectively, the closest value available for $\nu 2d_{5/2}$ is from the magnetic-dipole moment of ^{87}Kr [Kei95].

A χ^2_ν analysis indicates a small preference for either an $I = (1)$ or (2) assignment (both with $\chi^2_\nu \approx 2.3$; other choices with $\chi^2_\nu > 2.8$), however the g -factor analysis indicates a more mixed picture. Both $I = (2)$ and (3) can be described by 82%:18% and 96%:4% linear contributions from the $(\pi 1f_{5/2} \otimes \nu 2d_{5/2})$ and $(\pi 1f_{5/2} \otimes \nu 3s_{1/2})$ configurations, respectively. Although the suggestion of ^{83}Ga with a dominant $\pi 1f_{5/2}^3$ proton configuration from LSSM calculations [Als16] further strengthens their case, the close proximity of $I = (4)$ and (5) to $(\pi 1f_{5/2} \otimes \nu 2d_{5/2})$ does not unequivocally rule them out. Overall, with the exception of maybe $I = (1)$, no nuclear-spin assignment can be ruled out in this analysis and thus, hyperfine parameters and electromagnetic moments displayed for ^{82}Ga in Chapter 6.4 assume the $I = (2)$ assignment from Ref. [Che12a]. Again as for the case with the two long-lived ^{80}Ga states in Chapter 6.3.2, the rather-pure nature for the $I^\pi = 2^-$ g factor in Figure 6.11 is at odds with the mixed configuration

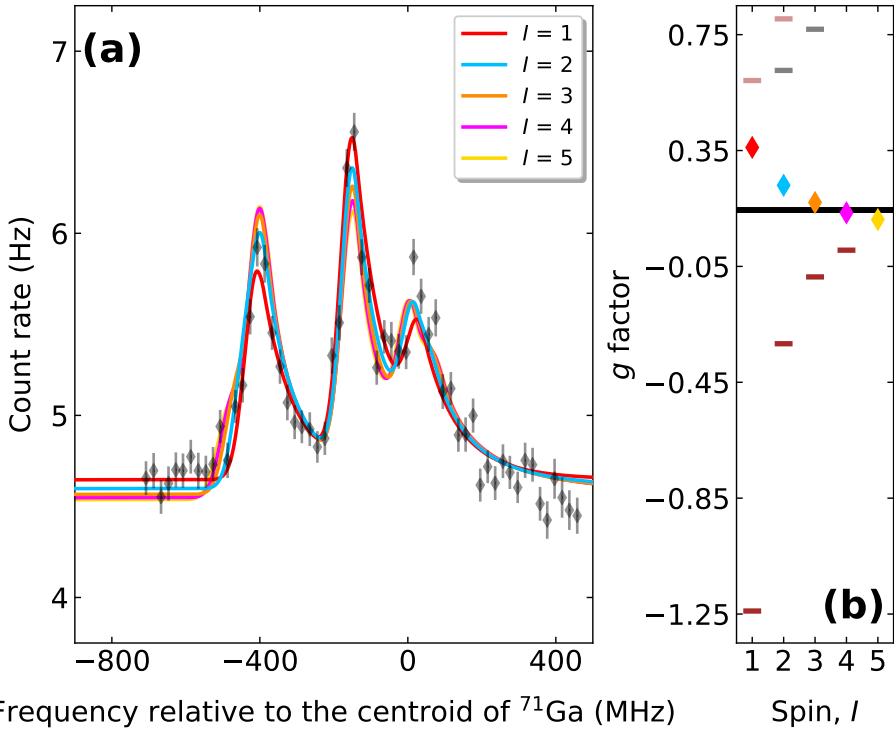


Figure 6.11: **(a)** The combined hyperfine spectrum of ^{82}Ga , fitted assuming $I = 1$ (red), 2 (blue), 3 (orange), 4 (magenta), and 5 (yellow) spin assignments. **(b)** Extracted g factors compared with applicable empirical values assuming pure: $(\pi 1f_{5/2} \otimes \nu 2d_{5/2})$ (solid-black line), $(\pi 1f_{5/2} \otimes \nu 3s_{1/2})$ (shaded-black lines), $(\pi 2p_{3/2} \otimes \nu 2d_{5/2})$ (solid-brown lines), and $(\pi 2p_{3/2} \otimes \nu 3s_{1/2})$ (shaded-brown lines) configurations.

predicted by the Paar-parabola analysis in Figure 6.6. once again highlighting the approximations made. Additionally, the only available $\nu 2d_{5/2}$ single-particle g factor from ^{87}Kr demonstrates the lack of measurements made past $N = 50$ in the ^{78}Ni vicinity; with $Z = 36$, it represents a rather poor choice considering the extra proton-pair correlations that are undoubtedly included in its g factor. The extraction and study of electromagnetic moments for $N = 51$ species, such as ^{81}Zn and ^{83}Ge , are thus crucial for further improving these calculations.

6.4 A summary of the hyperfine parameters and electromagnetic moments for $^{80,82}\text{Ga}$

A - and B -hyperfine parameters for $^{80g,80m}\text{Ga}$ and ^{82}Ga are presented in Tables 6.4 and 6.5. Following the theme for the other gallium measurements presented in this thesis, final magnetic-dipole and electric-quadrupole moments arise from a weighted mean of available hyperfine parameters involved in the 417.2-nm transition. Magnetic-dipole moment calculations for the states in ^{80}Ga involved the $A(4p\ ^2P_{3/2})$ parameters because this was the chosen extracted parameter in Ref. [Che10a]. Electromagnetic moments presented in these tables are displayed in Figure 6.12 alongside other even- A species in the vicinity.

Shell-model comparisons using the JUN45, jj44b, and fpg interactions, with the even- A gallium electromagnetic moments are provided in Figures 6.12(a) and 6.12(b). It is remarked that g -factor comparisons with fpg calculations are not possible because these were not provided in Ref. [Sri11]; neither is the jj44b electric-quadrupole calculation for ^{72}Ga because of the same reason in Ref. [Man11a]. With respect to the g -factor comparisons, both the JUN45 and jj44b calculations perform equally as well; there is a slight overall preference for JUN45 towards the neutron-rich side, and likewise for jj44b for the isotopes near $N = 40$. The best description for the electric-quadrupole moments is provided by the jj44b calculations, with those from the fpg interaction matching favourably; JUN45 does not perform well for the two long-lived states in ^{80}Ga . However for $N \leq 45$, there is a general breakdown in agreement concerning all three shell-model interactions, highlighting possible difficulties in being able to account for an increasing quantity of pn correlations towards the mid-shell. None of these interactions can venture across $N = 50$ towards ^{82}Ga because of the non-inclusion of the $\nu 2d_{5/2}$ orbital in any of the model spaces. Its inclusion in

Table 6.4: Extracted A -hyperfine parameters for $^{80g,80m,82}\text{Ga}$, compared with available literature values; COLLAPS subcolumns display data taken from Refs. [Che10a; Che12a]. Magnetic-dipole moments have been calculated from a weighted mean of available A -hyperfine parameters. Uncertainties are enclosed within parentheses.

A	I	$A(4p\ ^2P_{3/2})$ (MHz)		$A(5s\ ^2S_{1/2})$ (MHz)		μ (μ_N)
		CRIS	COLLAPS	CRIS	COLLAPS	Combined
80	6	+0.6(3)	+0.9(1)			+0.036(3)
80	3	-68.3(3)	-67.5(1)	-380.3(10)	-377.6(8)	-1.430(7)
82	(2)			+182.8(27)	+182.5(14)	+0.459(1)

Table 6.5: Extracted B -hyperfine parameters for $^{80g,80m,82}\text{Ga}$, compared with available literature values. Electric-quadrupole moments have been calculated from a weighted mean of available B -hyperfine parameters. Uncertainties are enclosed within the parentheses.

A	I	$B(4p^2P_{3/2})$ (MHz)		Q (efm ²)
		CRIS	Refs. [Che10a; Che12a]	Combined
80	6	+177(1)	+174(3)	+49.6(11)
80	3	+130(3)	+137(3)	+37.4(12)
82	(2)	+57(6)	+72(3)	+19.2(18)

the A3Da-m interaction have already helped to interpret the nuclear-structure nuances that arise in the electromagnetic moments of ^{29}Cu [Gro17b] and ^{30}Zn [Yan16b; Wra17] isotopes near $N = 50$. Thus, it is strongly suggested to explore this interaction for these gallium isotopes not only to improve on existing calculations, but also to provide theoretical values for ^{82}Ga and beyond.

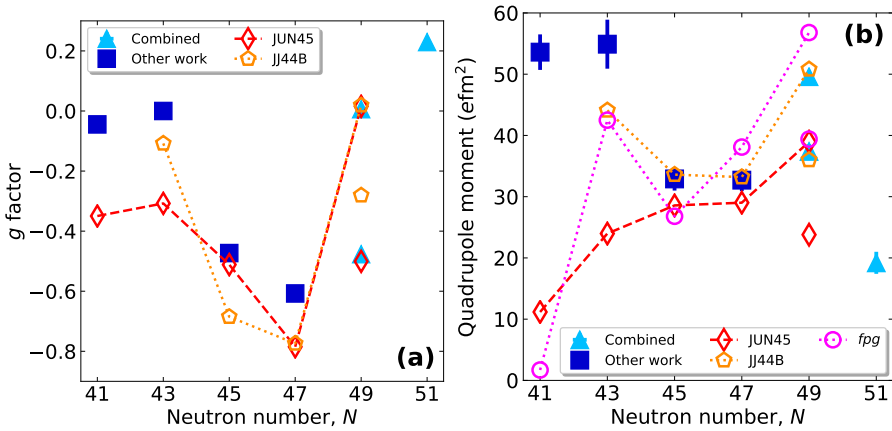


Figure 6.12: (a) Extracted g factors and (b) electric-quadrupole moments for even- A gallium isotopes from $N = 41 - 51$ inclusive. Values presented in this thesis (light-blue triangles) and other literature values (dark-blue squares; [Man11a]) are compared to JUN45 (red diamonds), jj44b (orange pentagons; [Man11a]) and fpg (pink circles; Ref. [Sri11]) calculations. Theoretical data points that are detached at $N = 49$ (for ^{80}Ga) represent calculations of the isomer state.

6.5 Isotope shifts and changes in the mean-square charge radii for $^{75,79-82}\text{Ga}$

Isotope-shift values for $^{75,79-82}\text{Ga}$ are presented in Table 6.6; in particular for $^{80g,80m}\text{Ga}$, their values reflect a combined data set from both the 2015 and 2017 experiments. Corresponding changes in the mean-square charge radii originate from a weighted mean of all available isotope-shift data, using the same atomic parameters in Ref. [Pro12] and **Article I**⁶. Although the isotope shifts reported in this thesis and in Ref. [Pro12] (not featured in Table 6.6) both possess systematic uncertainties related to the acceleration voltage, they are not directly comparable to one another; thus, the changes in the mean-square charge radii presented in the aforementioned table are not assigned with such an uncertainty. Nonetheless, the manifestation of these types of uncertainties in both data sets, which can be as significant as those borne from the statistics alone, serve as a strong motivation for the re-measuring of gallium isotope shifts in the future.

Supplemented with other changes in the mean-square charge radii for isotopes not measured at the CRIS experiment, Figure 6.13 displays the overall charge-radii landscape of the relevant isotope chains in the neighbourhood of ^{78}Ni . Reference charge-radii values for a particular chain have been taken from Ref. [Fri04]. Exclusively for isotope chains with multiple data sets available: final charge-radii values result from an isotope shift weighted mean if the same transition was involved (like for gallium), otherwise from a weighted mean of the charge-radii results. The most noticeable feature of the ^{31}Ga isotope chain is the distinctive kink seen past $N = 50$ which signals the crossing of a shell

Table 6.6: Isotope shifts and changes in the mean-square charge radii for $^{75,79-82}\text{Ga}$; see text for the evaluation details regarding the latter parameter. Statistical, modified-systematic, and atomic-systematic uncertainties are enclosed within parentheses, brackets, and braces respectively.

A	I	$\delta\nu^{71,A}$ (MHz)		$\delta\langle r^2 \rangle^{71,A}$ (fm ²)
		CRIS	Ref. [Pro12]	Combined
75	3/2	-47(1)[1]	-45(2)	+0.282(2){58}
79	3/2	-185(12)[2]	-186(2)	+0.291(1){87}
80	6	-245(5)[3]	-239(4)	+0.237(7){91}
80	3	-238(5)[3]	-232(3)	+0.257(6){92}
81	5/2	-266(3)[3]	-272(2)	+0.245(6){99}
82	(2)	-235(3)[3]	-222(9)	+0.428(11){118}

⁶ $M_{417} = -211.4(210)$ GHz amu, $F_{417} = +0.40(6)$ GHz/fm².

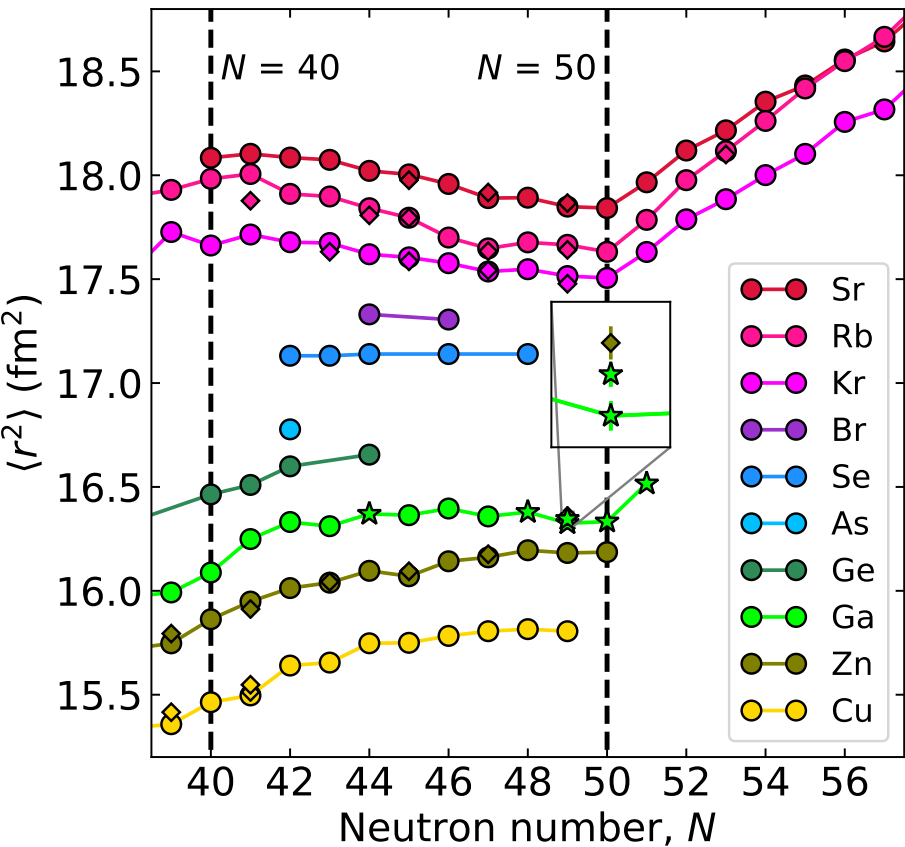


Figure 6.13: Mean-square charge radii for isotopes (ground and long-lived isomer states: circles and diamonds, respectively) in the vicinity of the ^{78}Ni region. Values for $^{75,79,80g,80m,81,82}\text{Ga}$ (taken from the ‘Combined’ column in Table 6.6) are represented with star markers. A 10x magnified region featuring (in ascending $\langle r^2 \rangle$): $^{80g,80m}\text{Ga}$, and ^{79m}Zn , is provided in the inset for better display. The $N = 40, 50$ (sub)shell closures are marked with dashed lines. References used for the compilation of this figure can be found in Appendix A.3.

closure [Cam16]. It is currently the lightest isotope chain in the vicinity of ^{78}Ni to exhibit this effect, since recent campaigns on those involving ^{29}Cu and ^{30}Zn did not manage to cross $N = 50$. Although this phenomena is encountered in the ^{36}Kr and ^{37}Rb isotope chains, both are relatively far away. This emphasises the need for not only measurements in isotope chains with scarce data, but also improved techniques for accessing neutron-richer ^{29}Cu and ^{30}Zn species.

Another noticeable feature is the gradual flattening of the charge radii starting from $N = 40$. This is also seen in ^{29}Cu , ^{30}Zn , ^{36}Kr , and ^{37}Rb , and could be interpreted as a sign of decreasing neutron collectivity approaching $N = 50$. Supplementary data from unmeasured isotope chains between ^{31}Ga and ^{36}Kr are needed in order to fully describe this interpretation.

6.6 Summary

In a region with relatively few experimental explorations, the study of neutron-rich species of gallium has been beneficial for further understanding the magical nature of the ^{78}Ni region. An examination of the g factors and electric-quadrupole moments of the odd- A family reveals an ever-changing nuclear structure, in which there is a gradual $\pi 2p_{3/2} \rightarrow \pi 1f_{5/2}$ migration as the $\nu 1g_{9/2}$ orbital is filled. While a complete proton transfer can be structurally observed ^{79}Ga with its seniority-3 ($\pi 1f_{5/2}^3$) $_{3/2^-}$ configuration, the $I^\pi = 3/2^- \rightarrow 5/2^-$ level actually manifests in ^{81}Ga based on a combined A -ratio and g -factor analysis. This behaviour is analogous to that seen in the ^{29}Cu isotope chain, however this study has not been able to confirm whether it persists for ^{83}Ga and beyond. Furthermore, the inconclusiveness of the A -hyperfine parameter ratio analysis for ^{81}Ga casts doubt on the confidence of being able to assign states possessing a high-value spin. Reiterating the statements made in Chapter 5.6, this further strengthens the need of measuring gallium under another transition which is more sensitive to the spin.

Further probing the magicity of $N = 50$ has also been achieved by the study of the even- A isotopes immediately before and after this shell closure. The presence of two long-lived low-lying states in ^{80}Ga is characteristic for an isotope belonging to the $N = 49$ isotone chain. Not only do their g factors correspond well to a $(\pi 1f_{5/2} \otimes \nu 1g_{9/2})$ configuration dictated by shell model predictions, they also match expectations from shell-model calculations. Even though ambiguity remains on the spin assignment for ^{80g}Ga , overall this provides additional assurance of the validity of shell-model principles. Additional $N = 50$ magicity is found with the kink in the mean-square charge radii value for the heaviest gallium isotope measured to date: ^{82}Ga . However, its condensed hyperfine structure complicates the nuclear-structure discussion: neither a firm spin assignment can be made, nor can the existence of additional isomeric structure be unequivocally confirmed or ruled out. The need for studying another transition in gallium outlined above is also applicable to ^{80g}Ga and ^{82}Ga , which would hopefully fully address these unresolved questions.

Chapter 7

Nuclear magicity near ^{208}Pb

With $Z = 82$ and $N = 126$, ^{208}Pb is the heaviest doubly-magic isotope that is currently accessible to experimental techniques. Many trans-lead nuclei can be

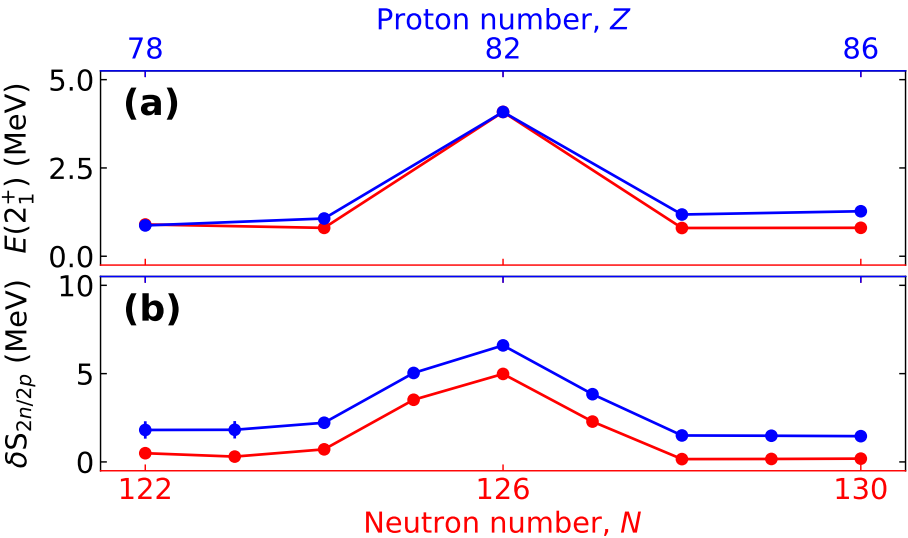


Figure 7.1: (a) Energies for the first excited 2^+ state. (b) Two-nucleon-separation energy differences between $N, N+2$ and $Z, Z+2$, for the neutrons and protons respectively. In effect, trends for $Z = 82$ (Pb) isotopes (red) and $N = 126$ isotones (blue) feature in both plots. Data are taken from Ref. [NND18].

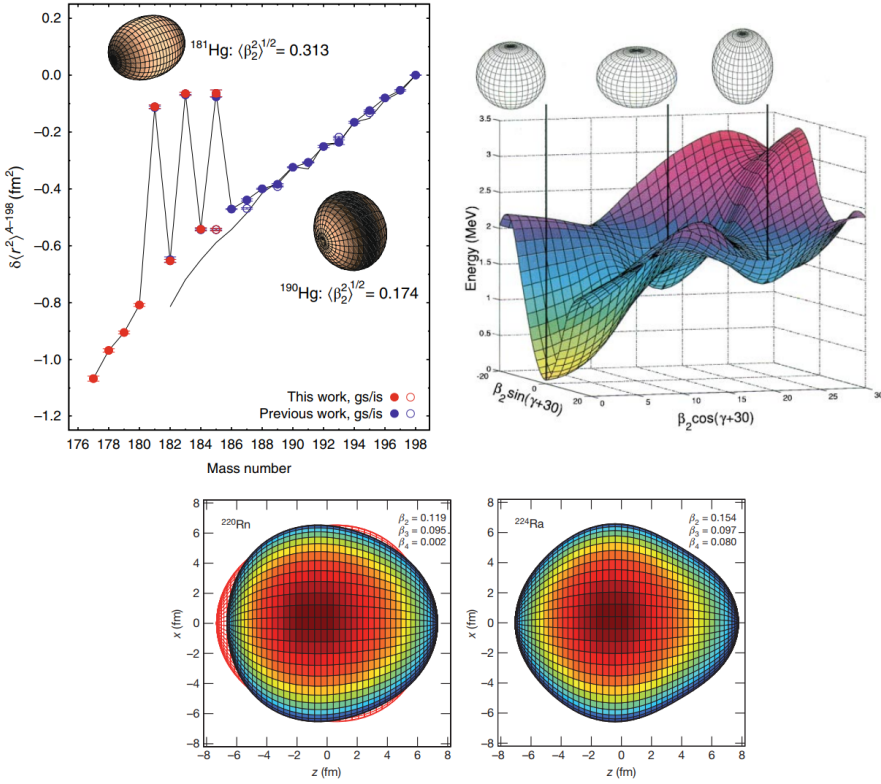


Figure 7.2: A trio of shape coexistence phenomena in the ^{208}Pb region. Pictures are taken from Refs. [Mar18a; And00; Gaf13] starting clockwise from the top-left. See text for further nuclear physics details.

produced with intense yields from UC_x targets, explaining why these isotopes are some of the most extensively studied over the past few decades at the ISOLDE facility [Bor17]. Trends observed for $E(2_1^+)$ and $S_{2n/2p}$ values across the isotope and isotone chains intersecting at ^{208}Pb in Figure 7.1 unequivocally indicates a doubly-magic character. Shape coexistence is a rather ubiquitous feature in this region: competing spherical and prolate ground-states for the most neutron-deficient $_{80}\text{Hg}$ isotopes [Ulm86; Mar18a], and a low-lying triplet of uniquely shaped $I^\pi = 0^+$ states in ^{186}Pb [And00] represent two examples that have attracted high-profile attention within the experimental-nuclear-physics community. Unconventional nuclear shapes have also been observed for ^{220}Rn and ^{224}Ra , exhibiting *pear* shapes because of octupole deformation [Gaf13].

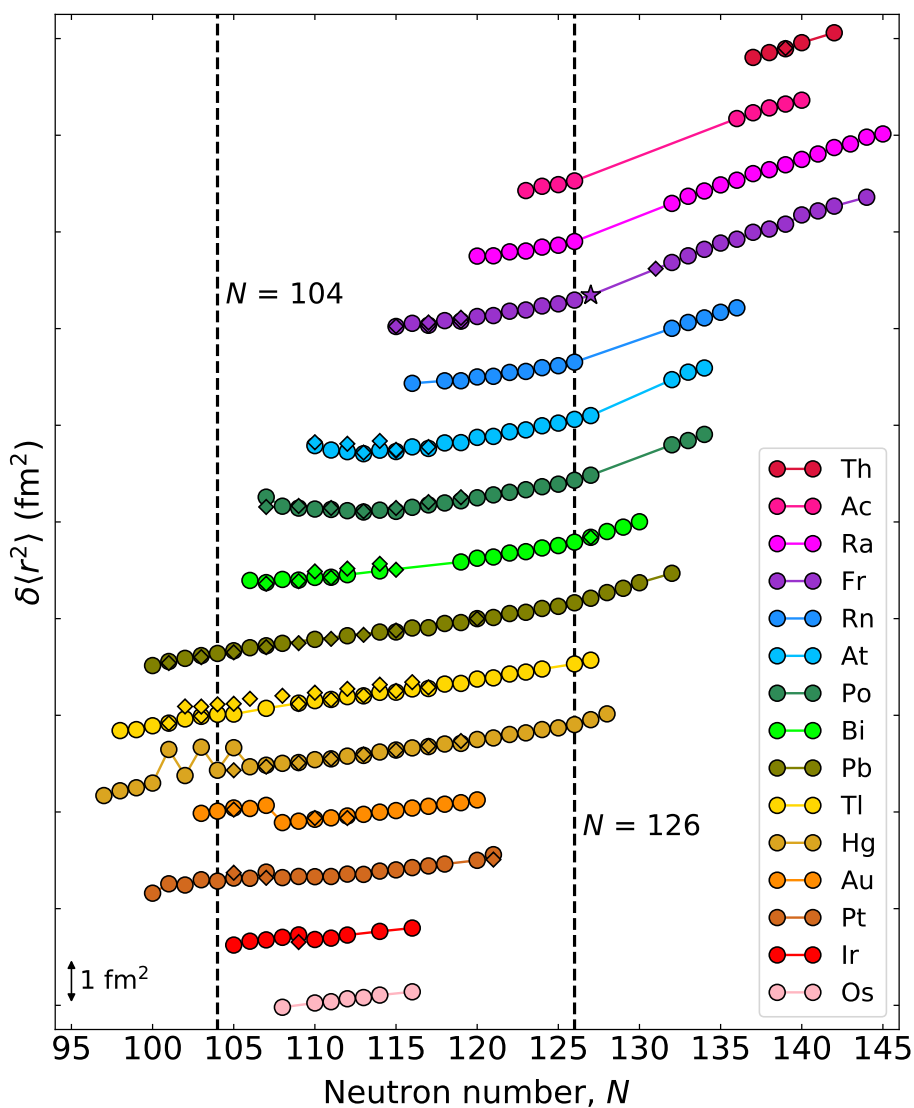


Figure 7.3: Changes in the mean-square charge radii for isotopes (ground and long-lived isomer states: circles and diamonds, respectively) in the vicinity of the ^{208}Pb region; offsets have been applied between isotope chains for a better display. The value for $^{214}\text{g}\text{Fr}$ is represented with a star marker, the measurements of which will be presented later in this chapter. The $N = 104$ mid-point and $N = 126$ shell closure are marked with dashed lines. References used for the compilation of this figure can be found in Appendix A.3.

Motivated by the surprising differences in the $\delta\langle r^2 \rangle$ values between $^{185,186}\text{Hg}$, there has been, and still is, a continuing effort to measure this parameter for all nearby isotope chains. A full up-to-date landscape of these values for ^{76}Os to ^{90}Th (at the time of writing this thesis) can be seen in Figure 7.3¹, in which the shape-coexistence manifestations² near the $N = 104$ subshell are visible. Some of the most desirable measurements however, are around $N = 126$ because they would crucially help to infer its magical character [Cam16]. Although distinctive increases in the $\delta\langle r^2 \rangle$ values, colloquially referred to as kinks, are observed immediately after $N = 126$ for ^{80}Hg , ^{81}Tl , ^{82}Pb , ^{83}Bi , and ^{84}Po , there is no theoretical consensus to explain the origin on these kinks. The SPE of the $\nu 2g_{9/2}$ and $\nu 1i_{11/2}$ orbitals form the basis of contention: while spin-orbit integration modifications to RMF calculations require a near-degeneracy to reproduce the kink in the ^{82}Pb and ^{84}Po isotope chains [Rei95; God13; Ebr16], other interactions use additional density-dependent terms derived from three-nucleon ($3N$) interactions without this requirement [Koh13; Nak15]. Thus in addition to desired $\delta\langle r^2 \rangle$ measurements for $N = 127$ isotopes, a complimentary g -factor measurement could reveal nuclear structure information that would help to provide further insight with respect to a correct theoretical picture. Even though the $\sim \mu\text{s}$ lifetime for most α emitters with $Z \geq 84$ and $N = 127 - 131$ is currently insurmountable for most laser-spectroscopy setups, $^{214g}_{87}\text{Fr}_{127}$ with a 5 ms half-life, is thankfully just-about accessible with these techniques.

The essence of **Article II** focuses on laser- and decay-spectroscopy measurements of $^{214g}_{87}\text{Fr}_{127}$. With five protons away from $Z = 82$ and a half-life of 5 ms, it not only represents the shortest-lived isotope to have been measured with laser spectroscopy techniques at an ISOL facility, but also the heaviest $N = 127$ isotone. Updated analyses and insights since its publication are provided in the successive sections to help further probe the magicity near ^{208}Pb .

7.1 Article II: Laser and decay spectroscopy of the short-lived isotope ^{214}Fr

N.B. Table II of this article has been modified into a double table in order to fit the dimensions of this document. No other aspect of this article has been changed.

¹Unlike the $\langle r^2 \rangle$ landscapes for ^{56}Ni and ^{78}Ni in Figures 5.9 and 6.13, a general lack of absolute charge-radii measurements for $Z \geq 84$ [Coc17] means that only the changes are displayed in Figure 7.3.

²Although not featuring in Figure 7.3, unpublished data for the ^{83}Bi isotope chain presented in Ref. [And17] shows the beginnings of a similar OES magnitude to that present in the ^{80}Hg isotope chain, at the same N number.

Laser and decay spectroscopy of the short-lived isotope ^{214}Fr in the vicinity of the $N = 126$ shell closure

G. J. Farooq-Smith,^{1,2,*} T. E. Cocolios,^{1,2} J. Billowes,¹ M. L. Bissell,^{1,2} I. Budinčević,² T. Day Goodacre,^{1,3} R. P. de Groote,² V. N. Fedosseev,³ K. T. Flanagan,¹ S. Franchoo,⁴ R. F. Garcia Ruiz,^{1,2} H. Heylen,² R. Li,^{4,†} K. M. Lynch,^{2,5} B. A. Marsh,³ G. Neyens,² R. E. Rossel,^{3,6} S. Rothe,^{1,3} H. H. Stroke,⁷ K. D. A. Wendt,⁶ S. G. Wilkins,¹ and X. F. Yang²

¹*School of Physics and Astronomy, The University of Manchester,
Manchester M13 9PL, UK*

²*KU Leuven, Instituut voor Kern- en Stralingsfysica, Celestijnenlaan 200D,
3001 Leuven, Belgium*

³*EN Department, CERN, CH-1211 Geneva 23, Switzerland*

⁴*Institut de Physique Nucléaire d'Orsay, F-91406 Orsay, France*

⁵*ISOLDE, PH Department, CERN, CH-1211 Geneva 23, Switzerland*

⁶*Institut für Physik, Johannes Gutenberg-Universität, D-55128 Mainz, Germany*

⁷*Department of Physics, New York University, New York, New York 10003, USA*

Abstract

Combined decay and laser spectroscopy measurements of the isotope ^{214}Fr are reported, using the collinear resonance ionization spectroscopy (CRIS) technique at the ISOLDE facility at CERN. For the $I^\pi = (1^-)$ spin assignment, the g -factor value of $g(^{214}\text{Fr}) = +0.241(16)$ corresponds to a relatively pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ ground-state configuration. An alternative interpretation with $g(^{214}\text{Fr}) = +0.144(10)$, for a (2^-) spin assignment suggests a greater contribution of configuration mixing with the $\nu 1i_{11/2}$ orbital. As the $N = 126$ shell closure is passed, a kink is observed at the $\delta\langle r^2 \rangle^{213,214}$ value, which is analogous to the behavior observed in the neighboring isotopic chains.

* gregoryjames.farooqsmith@kuleuven.be

† Present address: TRIUMF, Vancouver, British Columbia V6T 2A3, Canada

I. INTRODUCTION

As one of the first elements studied at the isotope separator online device (ISOLDE) facility at CERN [1], francium has played an important role in explaining the variety of nuclear behavior that exists around the $Z = 82$ and $N = 126$ magic shell closures. High-profile studies have already documented the phenomena of shape coexistence and octupole deformations in isotopes located in the neutron-deficient and -rich regions, respectively [2, 3]. However for the case of $^{214}_{87}\text{Fr}_{127}$, which resides between these two regions next to the $N = 126$ shell closure, little information is known. In this paper we focus on the charge radii in the trans-lead region and in particular, the characteristic increase in the gradient that is observed at a shell closure crossing. This is often referred to as a “kink” [4] and has already been seen at $N = 126$ in the neighboring isotopes from thallium to radium [5–12].

The interpretation of the kink at $N = 126$ remains a challenge for nuclear models. Many groups have investigated this region with mean-field calculations [13–17] but no general consensus exists on how to reproduce the correct magnitudes. Until now, limited experimental input has been available regarding the gradient of the kink at this particular shell closure. Although data for the trans-lead region is available from $N = 132$ onwards, little is known for $N = 127 - 131$ due to the short lifetime of the isotopes immediately after the $N = 126$ shell closure. Only four $N = 127$ isotones have been studied: $_{81}\text{Tl}$ [5], $_{82}\text{Pb}$ [6], $_{83}\text{Bi}$ [7] and $_{84}\text{Po}$ [18]. With 5 protons located in the $\pi 1h_{9/2}$ orbital, ^{214}Fr offers an additional test of the robustness of the $Z = 82$ shell closure.

The collinear resonance ionization spectroscopy (CRIS) setup at CERN-ISOLDE has recently explored isotopes across the francium chain [11, 19–22]. In particular, new developments at the decay spectroscopy station (DSS) [23] have allowed combined decay and laser spectroscopy techniques to be performed. A recent example of this includes the identification of ground and isomeric states in ^{202}Fr , ^{204}Fr , and ^{206}Fr [20, 22], from overlapping hyperfine structures. In this article, we report the first measurements of ^{214}Fr at the CRIS beam line. With a half-life of only 5.0(2) ms [24], ^{214}Fr represents the shortest-lived isotope to have been measured online with laser spectroscopy. The change in the mean-square charge-radii and the gyromagnetic factor will be presented, alongside a discussion on recent mean-field calculations performed in the area. Based on these considerations, the implications on the behavior exhibited past the $N = 126$ shell closure will be discussed.

II. EXPERIMENTAL SETUP

The ^{214}Fr ion beam was produced at the ISOLDE facility [25] via spallation reactions, by impinging 1.4 GeV protons from the proton-synchrotron booster (PSB) onto a thick UC_x target [26]. The PSB duty cycle delivered proton pulses separated by integral multiples of 1.2 seconds, resulting in release profiles which could be defined empirically [27]. The short half-life of ^{214}Fr meant that the release profile was primarily dominated by the decay losses. This increased the importance of operating the experimental setup at a fast duty cycle comparable to the half-life, in order to minimize these losses.

The resulting reaction products were surface-ionized in a high temperature (~ 2200 K) tantalum hot cavity ion source and extracted at 40 keV. The isotopes of interest were mass separated by the high resolution mass separator (HRS) [25]. Cooling and bunching of the ions was performed with the use of the ISCOOL radio-frequency gas-filled linear Paul trap [28, 29]. The release time for these bunches was synchronized with the duty cycle of the laser system. In order to limit the ^{214}Fr decay losses from within the trap, a 200 Hz repetition rate was used. After reaccelerating to 40 keV, the ion bunches were electrostatically deflected into the CRIS beam line [20, 30, 31], where they were first neutralized through a charge exchange cell (CEC) filled with potassium vapor (445 K – 455 K) with a neutralization efficiency of $\sim 50\%$. Any remaining nonneutralized ions were deflected out of the atomic beam in the differential pumping region situated just after the CEC.

The neutralized atomic bunches then entered an ultra high vacuum (UHV) interaction region (kept at a pressure of 9×10^{-9} mbar), where they were collinearly overlapped with laser light both spatially and temporally. The ionization scheme consisted of two steps: a 422.7-nm step to excite the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition at $23\ 658.306\ \text{cm}^{-1}$, and a nonresonant 1064-nm step which was subsequently used to ionize the atoms. The 422.7-nm laser light was produced by frequency-doubling light from a 10 kHz Ti:Sa narrowband (1 GHz) laser provided by the resonance ionization laser ion source (RILIS) laser laboratory [32, 33], in which the laser frequency could be stepped by using a data acquisition routine [34]. It was transported to the CRIS beam line using a 35-m long multimode optical fiber. The 1064-nm light was produced with a Litron LPY-601 Nd:YAG system (consisting of two lasers running at 100 Hz each) that could operate at 200 Hz, situated next to the CRIS beam line. By opening the beamgate 10 ms after proton impact with the target and setting

its temporal width to several ^{214}Fr half-lives, the amount of ^{214}Fr available for study was maximized. This also limited the isobaric contamination present (mainly ^{214}Ra), which was constantly released from the target. The low pressure maintained within the interaction region minimized the background due to nonresonant ionization with gas molecules. After the resonant excitation and reionization processes, the ions were deflected onto a biased copper plate (held at -270 V), where ejected secondary electrons were guided onto a micro-channel plate (MCP) detector. Separate α -decay spectroscopy could also be performed by implanting the ions into carbon foils on a rotatable wheel. Emitted α particles could be detected within the DSS [20, 23], located at the end of the CRIS beam line.

III. RESULTS

A. Laser Spectroscopy of ^{214}Fr

The magnetic dipole moment was extracted from the hyperfine structure shown in Fig. 1 by means of a χ^2 -minimization fitting routine [4]. The center of gravity of the hyperfine spectrum of ^{214}Fr was compared to the reference isotope, ^{221}Fr , for extraction of the isotope shift. All other measured observables were determined relative to ^{210}Fr , using: $A_{\text{ref}}(7s^2S_{1/2})$

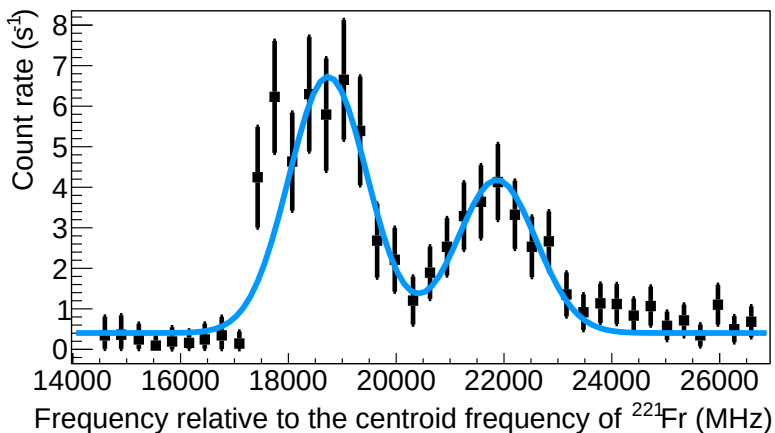


FIG. 1. Collinear resonance ionization spectroscopy of ^{214}Fr . Each data point represents an average of 72 s of data recording.

$= +7195.1(4)$ MHz [35], $I^\pi = 6^+$ [36], and $\mu_{\text{ref}} = +4.38(5) \mu_N$ [37]. The changes in the mean-square charge radii were extracted by using the equation

$$\delta\nu^{AA'} = M_{422} \frac{A' - A}{AA'} + F_{422} \delta\langle r^2 \rangle^{AA'}, \quad (1)$$

where A and A' are assigned to the reference and measured isotopes respectively. The parameters $M_{422} = 750(330)$ GHz.amu and $F_{422} = -20.67(21)$ GHz/fm² represent the atomic factors and contain all the optical dependence for the 422.7-nm transition [20]. The hyperfine anomaly for francium has been measured as $<1\%$ [38, 39], which was beyond the resolution of the technique used in this work. The data were analyzed with a constant ratio of $A(7s^2S_{1/2})/A(7p^2P_{3/2}) = +0.0036$ and the $B(7p^2P_{3/2})$ hyperfine parameter was set to zero [30]. The 1.5-GHz linewidth of the 422.7-nm RILIS Ti:Sa laser produced pure Gaussian line shapes. Only the lower-state splitting could be resolved, preventing the extraction of the nuclear spin and quadrupole moment. However, the comparison of measured peak intensity ratios with those expected from angular-momentum coupling meant that tentative spin assignments were possible. The low resolution available, along with $J = 1/2$ for the $7s^2S_{1/2}$ state, meant that the normal relation for the hyperfine transition intensities, $S_{FF'}$, between states of F and F' (with angular momentum J and J' respectively) could be simplified to

$$\frac{S_{FF'}}{S_{JJ'}} = \frac{I+1}{I}, \quad (2)$$

where I represents the nuclear spin of the isotope. Fitting routines applied to the data shown in Fig. 1 suggested a ratio of 1.7(3) for the intensities of the two structures. In conjunction with Eq. (2), this was consistent with a low-spin assignment. In order to assess the ratio of ground and isomeric states within the beam and their contribution to the structure of Fig. 1, additional decay spectroscopy measurements were performed.

A systematic error of 30 MHz was established for $A(7s^2S_{1/2})$ based on the scatter observed in the measured A factors of the ^{221}Fr reference scans [11, 30]; however, this uncertainty was mainly dominated by the limited resolution of the laser system. For the isotope shift, a minimum instrument error of 150 MHz was set. This accounted for the drift and fluctuations of the centroid frequency of ^{221}Fr , and the precision of the HighFinesse/Ångstrom WS7 wavelength meter. When the calculated weighted standard deviation of the isotope shift or $A(7s^2S_{1/2})$ parameter was greater than the minimum errors, these were quoted instead.

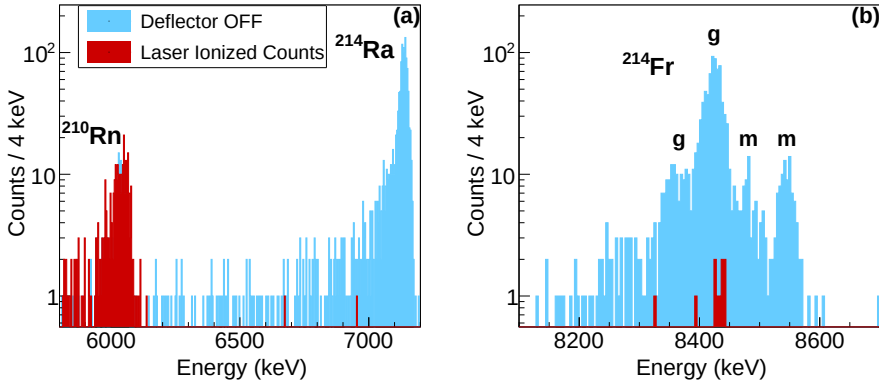


FIG. 2. α -decay energy spectra showing the content of the full (blue) and laser-ionized (red) ^{214}Fr beams at the DSS. (a) An α -decay energy spectrum for ^{214}Ra ($E_\alpha = 7137(3)$ keV, $t_{1/2} = 2.46(3)$ seconds [40]) and its daughter ^{210}Rn ($E_\alpha = 6041(3)$ keV, $t_{1/2} = 2.4(1)$ h [41]). (b) An α -decay energy spectrum for ^{214g}Fr and ^{214m}Fr ; lifetimes and decay energies can be found in Fig. 3.

B. Decay Spectroscopy of ^{214}Fr

Laser-assisted nuclear decay spectroscopy was performed for the ^{214}Fr ion beam in order to determine its ground and isomeric state content. Figures 2(a) and 2(b) show the α -decay spectra from the implantation of the full $A = 214$ beam, as well as the purified laser-ionized beam of ^{214}Fr into the carbon foils, for a collection time of 30 min. Relative to the centroid frequency of ^{221}Fr , the Ti:Sa laser frequency was detuned to 18.3 GHz and 22.1 GHz, respectively, for each hyperfine structure observed in Fig. 1. The laser-ionized spectrum in Fig. 2(b) represents the sum of the two separate spectra obtained from this method. A selectivity of 3.3 [20] meant that the content of each peak could be analyzed separately at the DSS. The presence of ^{214}Ra as an isobaric contaminant was reduced from the laser-ionized spectrum by beam gating (see Sec. II). However, the impinging of its daughter isotope ^{210}Rn ($t_{1/2} = 2.4(1)$ h [41]) with the surface of the silicon detectors occurred whenever full ^{214}Fr beams were examined. This meant that its decay could still be seen when analyzing the laser-ionized beam, allowing the ^{210}Rn peak to be used for calibrating the energy scale for both spectra. All expected α -decays from Fig. 3 were observed for both ^{214g}Fr and ^{214m}Fr when the ion deflector after the CEC was switched off, with a ratio of approximately

8:1. However, the implantation of the laser-ionized beam resulted in lower statistics and a nonoptimal energy resolution for the silicon detectors, due to the influence of the laser light on the baseline of the detector signal. In addition, data taking for this measurement was stopped before it could be completed (as this was performed at the end of the experiment), resulting in only a partial data set being obtained. Nonetheless, 10 counts were observed which corresponded to the decay of ^{214}gFr , and none were seen for $^{214\text{m}}\text{Fr}$. Given the observed $^{214}\text{gFr};^{214\text{m}}\text{Fr}$ ratio of 8:1 with the full beam, a statistical analysis concluded there was a 100% confidence that the ground state was present in the laser-ionized beam. Additionally, there was an 82.5% confidence that no isomeric state was present in the laser-ionized beam. Although the possibility of a ground/isomeric state mix could not be ignored *a priori*, any

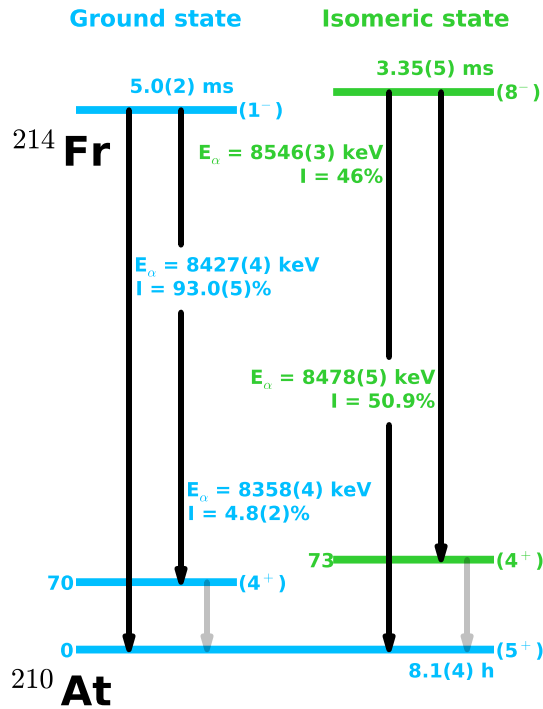


FIG. 3. (Color on-line) The radioactive decay scheme of $^{214\text{g,m}}\text{Fr}$. The solid arrows represent the main α -decay paths [40] and the shaded arrows depict the allowed γ -ray transitions between given states.

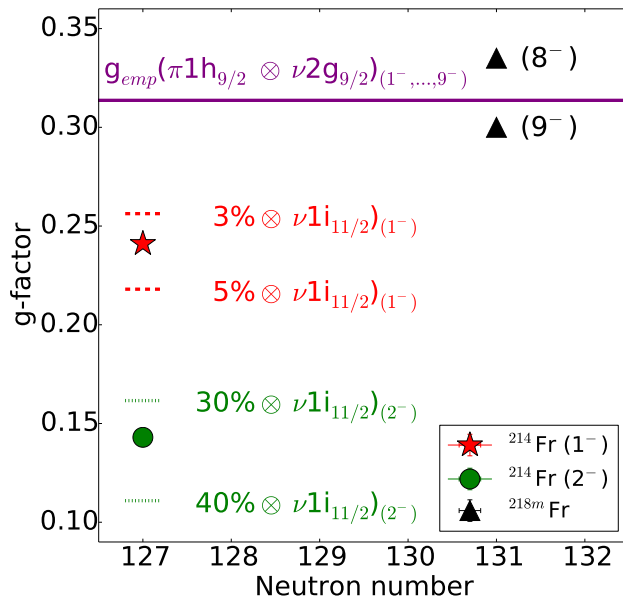


FIG. 4. (Color on-line) Extracted ^{214}gFr g factors for (1^-) and (2^-) spin assignments, alongside those for the tentative assignments of $^{218\text{m}}\text{Fr}$ [11]. Associated error bars are smaller than the data points. The g factors for a pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration and those with $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ configuration mixing have been calculated empirically. See text for details.

existence of an isomeric state in Fig. 1 would have been suppressed by a factor of eight with respect to the ground state. This was consistent with background, meaning the isomeric state contribution could be neglected.

IV. DISCUSSION

To first order, odd-odd ^{214}gFr involves the coupling of an unpaired valence proton located in the $\pi 1h_{9/2}$ orbital with a single valence neutron above the $N = 126$ shell gap, where $\nu 2g_{9/2}$ is the next spherical shell model orbital. This leads to possible ground state spin assignments of $(0, 1, 2, \dots)^-$, considering the lowest spin states from this multiplet. The observed hyperfine structure of the ground state in Fig. 1 automatically rules out a $I = (0^-)$ spin assignment, due to the observation of two peaks. Therefore, the use of $I = (1^-)$ and $I = (2^-)$ low-spin

assignments were considered for the laser spectroscopy analysis following the observations presented above. Earlier α -decay spectroscopy studies have also ruled out the (0^-) spin assignment and suggest a (1^-) spin/parity, based on analogy with the isotone ^{210}Bi [42–44]. The extracted g -factor values for the (1^-) and (2^-) spin assignments are presented in Table I and shown graphically in Fig. 4. The calculation of the expected empirical g factor (represented by the purple solid line in Fig. 4) can be obtained from the additivity relation [45]. For the case where the proton and neutron spins are the same, the g factor of the weakly coupled pn configuration is independent of the spin to which they couple to. Individual empirical g factors for $\pi 1h_{9/2}$ and $\nu 2g_{9/2}$ were taken from the magnetic moments of the closest odd- A isotopes available: ^{213}Fr [35] and ^{211}Po [46], respectively.

Additionally, the $\nu 1i_{11/2}$ and $\nu 2g_{7/2}$ orbitals are also in the near vicinity [15] and their contributions to the total g factor due to configuration mixing with the $\nu 2g_{9/2}$ orbital need to be considered. With respect to the $\nu 1i_{11/2}$ orbital, only linear contributions are expected and this is reflected in Fig. 4. The experimental $g(^{214}\text{gFr}_{I=(1^-)}) = +0.241(16)$ is consistent with a relatively pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})_{1^-}$ configuration. It also compares to the g factors obtained for the tentative $I = (8^-)$ or $I = (9^-)$ assignments of $^{218\text{m}}\text{Fr}$ at $N = 131$, which is suggested to have a similar single-particle configuration with the protons and neutrons aligned [11]. The contribution due to configuration mixing with the $\nu 1i_{11/2}$ orbital (using the calculated magnetic moment for ^{209}Pb [47]) is relatively small, at 3.8%. However in contrast, $\nu 1i_{11/2}$ configuration mixing accounts for 33.5% of the ^{214}Fr ground state assignment when $I = (2^-)$, with $g(^{214}\text{gFr}_{I=(2^-)}) = +0.144(10)$. In either case, evidence of a contribution from the $\nu 1i_{11/2}$ orbital supports previous theoretical studies that require this in order to reproduce the kink in the mean square charge-radii values at the $N = 126$ shell closure [14, 15, 17]. For

TABLE I. The $A(7s\ ^2S_{1/2})$ factors, isotope shifts, g factors, and the change in the mean-square charge-radii for ^{214}Fr with respect to ^{221}Fr , for assumed (1^-) and (2^-) spin assignments. The statistical and systematic uncertainties for $\delta\langle r^2 \rangle^{214,221}$ are separated, enclosed within parentheses and brackets, respectively.

I^π	$A(7s\ ^2S_{1/2})$ (GHz)	$\delta\nu^{221,214}$ (GHz)	g factor	$\delta\langle r^2 \rangle^{221,214}$ (fm ²)
(1^-)	+2.37(15)	+19.51(18)	+0.241(16)	−0.949(9){10}
(2^-)	+1.42(9)	+19.77(18)	+0.144(10)	−0.962(9){10}

TABLE II. Mean square charge-radii values just before and after the $N = 126$ shell closure for Hg to Ra, using Eq. (3) and literature mean square charge-radii values. The Fr $\delta\langle r^2 \rangle^{126,127}$ value corresponds to a (1^-) assignment and entries are marked with a ‘-’ where data is currently unavailable. Only statistical uncertainties are quoted for these values.

	Hg	Tl	Pb	Bi	
$\delta\langle r^2 \rangle^{125,126} \text{ (fm}^2\text{)}$	0.0654(25)	-	0.0720(1)	0.0710(22)	
$\delta\langle r^2 \rangle^{126,127} \text{ (fm}^2\text{)}$	-	0.0782(130)	0.0910(5)	0.0990(34)	
Refs.	[48]	[5]	[6]	[7]	
	Po	At	Rn	Fr	Ra
$\delta\langle r^2 \rangle^{125,126} \text{ (fm}^2\text{)}$	0.0813(12)	-	0.0735(1)	0.0790(1)	0.0773(39)
$\delta\langle r^2 \rangle^{126,127} \text{ (fm}^2\text{)}$	0.1040(12)	-	-	0.1056(92)	-
Refs.	[18]	-	[9]	This work, [10]	[49]

the $\nu 2g_{7/2}$ orbital, mixing calculations have been performed using the same empirical values as mentioned above for the $\pi 1h_{9/2}$, $\nu 2g_{9/2}$, and $\nu 2g_{7/2}$ orbitals, as well as the calculated $B(M1)$ value connecting the $\nu 2g_{9/2}$ and $\nu 2g_{7/2}$ orbitals [47]. These have shown that a 3.8% admixture of this type is sufficient to describe the measured g factor for ^{214}Fr in the case of $I = (1^-)$. For the $I = (2^-)$ assignment, however, the $\nu 2g_{7/2}$ orbital cannot contribute to the total g factor. Further study is required to confirm the ground-state spin and to investigate any possible shell effects that might be present, which will serve to test the robustness of the $N = 126$ shell closure.

The change in the mean-square charge-radii is interpreted in Fig. 5. Comparing the difference in the mean-square charge-radii before and after $N = 126$ offers an alternative way of isolating the effect seen at $N = 126$. This difference can be calculated by using

$$\delta\langle r^2 \rangle^{N,N'} = \delta\langle r^2 \rangle^{N,M} + \delta\langle r^2 \rangle^{M,N'}, \quad (3)$$

with N, N' representing the neutron numbers 125, 126 and 126, 127, for the differences before and after the shell closure respectively. The values for Hg-Ra are given in Table II and neutron numbers are used in the mean-square charge-radii superscripts to highlight the crossing of the $N = 126$ shell closure. A first observation is that the gradient in the charge radii between the $N = 125$ and $N = 126$ isotones is smaller than the gradient between $N =$

126 and $N = 127$. The latter leads to the well-known kink in the radii values [4]. A second observation is that the change in the radii below $N = 126$ is more or less constant for all isotopes from Hg ($Z = 80$) up to Ra ($Z = 88$), while an increase of the slope in the kink is observed with an increasing number of protons beyond $Z = 82$. It should be noted that for the cases of ^{83}Bi and ^{84}Po , large systematic uncertainties on the atomic parameters could affect this interpretation [50]. However, in accordance with Eq. (1) these uncertainties scale with the mass difference. Therefore, the close proximity of the charge-radii values in Fig. 5 with their references minimizes these systematic effects. Despite this, the francium data points for both spin assignments lie far away from this trend. Further studies on ^{214}Fr in higher resolution is needed to confirm this result and draw conclusions about the influence of the valence protons outside $Z = 82$ on the slope of the kink. Additionally, data from the

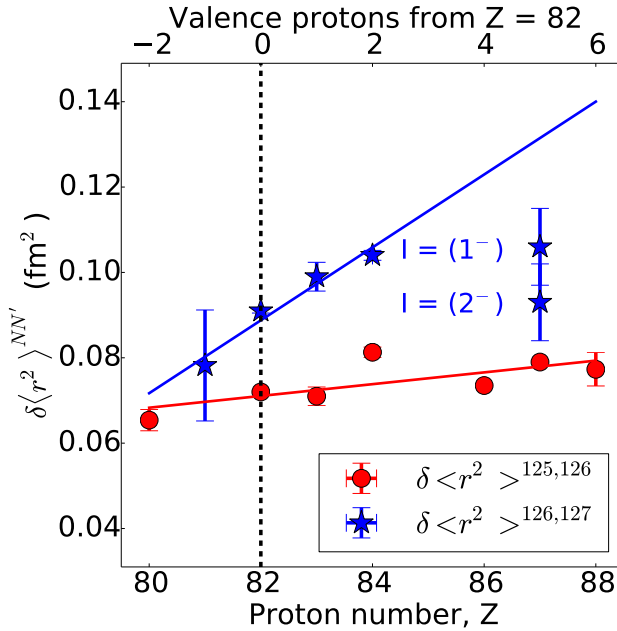


FIG. 5. (Color on-line) The difference in the mean-square charge radii for before (red circles) and after (blue stars) the $N = 126$ shell closure, across the $N = 125$ and $N = 127$ isotone chains, using the values in Table II. The shell closure at $Z = 82$ is highlighted with a dashed line.

$_{80}\text{Hg}$, $_{85}\text{At}$, $_{86}\text{Rn}$, and $_{88}\text{Ra}$ isotones are needed in order to fully establish the systematics in this region.

The question of how this mechanism arises is still debated. Modifications to the spin-orbit interaction in mean-field calculations have successfully reproduced the kink in the Pb and Po isotope chains [14, 15, 17]. Other studies suggest the inclusion of a density-dependent term to the spin-orbit term [13, 16], with the latter also proposing the inclusion of three-nucleon ($3N$) interactions in mean-field calculations. However, mean-field theory is primarily limited to even-even nuclei. Such calculations are difficult to perform for nuclei with odd numbers of protons and neutrons such as $^{214}_{87}\text{Fr}_{127}$, and this poses a major challenge for theoretical nuclear physics [51]. Recently, modified mean-field methods have allowed calculations to extend to odd- A nuclei, giving reasonable descriptions to light-mass nuclei such as ^{25}Mg [52]. Density Functional Theory (DFT) has also been developed in recent years and it is hoped that this can be applied across the entire nuclear landscape [53]. At present, a limited framework exists for mean-field calculations in this area of the nuclear chart and the mechanism remains an open question. Further studies are required to expand on the current nuclear theory constraints and it is anticipated that future developments of DFT calculations can be extended to include calculations for heavy-mass nuclei. Experimentally, the study of other $N = 127$ isotones is desirable in order to further assert whether a dependence exists between the magnitude of these kinks and the proton number.

V. CONCLUSION

The neutron-deficient isotope ^{214}Fr has been studied at the CRIS beam line at ISOLDE. With a half-life of 5.0(2) ms, ^{214}gFr represents the shortest-lived isotope studied online with laser spectroscopic techniques. A comparison between empirical and experimentally obtained g factors suggest that whilst the possible $I = (1^-)$ assignment is dominated by a $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration, the alternative $I = (2^-)$ assignment involves more configuration mixing with the $\nu 1i_{11/2}$ orbital. Further investigation into possible shell effects involving this orbital is required for a definitive assignment. The charge-radii value past the $N = 126$ shell closure shows the expected kink and is comparable to the behavior exhibited in the neighboring isotones.

VI. ACKNOWLEDGEMENTS

We acknowledge the support of the ISOLDE collaboration and technical teams, and the GSI target laboratory for producing the carbon foils. This work was supported by the IUAP-Belgian State Belgian Science Policy (BRIX network P7/12), FWO-Vlaanderen (Belgium) and GOA 015/10 from KU Leuven; ERC Consolidator Grant No.648381; the Science and Technology Facilities Council Consolidated Grant No.ST/F012071/1, Continuation Grant No.ST/J000159/1 and Ernest Rutherford Grant No.ST/L002868/1; and the European Union's Seventh Framework Programme for Research and Technological Development under Grant Agreements No. 262010 (ENSAR), No. 267194 (COFUND), and No. 289191 (LA³NET). T. E. C. was supported by the STFC Ernest Rutherford Fellowship No.ST/J004189/1. K. T. F. was supported by STFC Advanced Fellowship Scheme Grant No.ST/G006415/1. K. M. L. was supported by the FWO Pegasus Marie Curie Fellowship under Grant No.267216. We acknowledge the financial aid from the Ed Schneiderman Fund at New York University.

-
- [1] P. G. Hansen *et al.*, Phys. Lett. B **28**, 415 (1969).
 - [2] A. N. Andreyev *et al.*, Nature **405**, 430 (2000).
 - [3] L. P. Gaffney *et al.*, Nature **497**, 199 (2013).
 - [4] P. Campbell, I. D. Moore, and M. R. Pearson, Prog. Part. Nucl. Phys. **86**, 127 (2016).
 - [5] 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, Phys. Rev. C **88**, 024315 (2013).
 - [6] R. C. Thompson *et al.*, J. Phys. G **9**, 443 (1983).
 - [7] M. R. Pearson, P. Campbell, K. Leerunnavarat, *et al.*, J. Phys. G **26**, 1829 (2000).
 - [8] 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. Fedosyeyev, 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, Phys. Rev. Lett. **106**, 052503 (2011).

- [9] W. Borchers *et al.*, *Hyperfine Interact.* **34**, 25 (1987).
- [10] V. A. Dzuba, W. R. Johnson, and M. S. Safronova, *Phys. Rev. A* **72**, 022503 (2005).
- [11] I. Budinčević, J. Billowes, M. L. Bissell, T. E. Cocolios, R. P. de Groote, S. De Schepper, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, K. M. Lynch, B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt, *Phys. Rev. C* **90**, 014317 (2014).
- [12] S. A. Ahmad *et al.*, *Phys. Lett. B* **133**, 47 (1983).
- [13] M. M. Sharma, G. Lalazissis, J. Konig, and P. Ring, *Phys. Rev. Lett.* **74**, 3744 (1995).
- [14] P. G. Reinhard and H. Flocard, *Nucl. Phys. A* **584**, 467 (1995).
- [15] P. M. Goddard, P. D. Stevenson, and A. Rios, *Phys. Rev. Lett.* **110**, 032503 (2013).
- [16] H. Nakada and T. Inakura, *Phys. Rev. C* **91**, 021302(R) (2015).
- [17] J. P. Ebran, A. Mutschler, E. Khan, and D. Vretenar, *Phys. Rev. C* **94**, 024304 (2016).
- [18] M. D. Seliverstov, T. E. Cocolios, W. Dexters, *et al.*, *Phys. Lett. B* **719**, 362 (2013).
- [19] K. T. Flanagan, K. M. Lynch, J. Billowes, M. L. Bissell, I. Budinčević, T. E. Cocolios, R. P. de Groote, S. De Schepper, V. N. Fedosseev, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt, *Phys. Rev. Lett.* **111**, 212501 (2013).
- [20] K. M. Lynch, J. Billowes, M. L. Bissell, I. Budinčević, T. E. Cocolios, R. P. De Groote, S. De Schepper, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt, *Phys. Rev. X* **4**, 011055 (2014).
- [21] 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, *Phys. Rev. Lett.* **115**, 132501 (2015).
- [22] K. M. Lynch, T. E. Cocolios, J. Billowes, M. L. Bissell, I. Budinčević, T. Day Goodacre, R. P. de Groote, G. J. Farooq-Smith, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, R. Li, B. A. Marsh, G. Neyens, R. E. Rossel, S. Rothe, H. H. Stroke, K. D. A. Wendt, S. G. Wilkins, and X. Yang, *Phys. Rev. C* **93**, 014319 (2016).
- [23] M. M. Rajabali, K. M. Lynch, T. E. Cocolios, *et al.*, *Nucl. Instrum. Methods A* **707**, 35 (2013).
- [24] S. C. Wu, *Nucl. Data Sheets* **110**, 681 (2009).

- [25] E. Kugler, *Hyperfine Interact.* **129**, 23 (2000).
- [26] A. Herlert, *Nucl. Phys. News* **20**, 5 (2010).
- [27] J. Lettry *et al.*, *Nucl. Instrum. Methods B* **126**, 130 (1997).
- [28] H. Frånberg *et al.*, *Nucl. Instrum. Methods B* **266**, 4502 (2008).
- [29] E. Mané *et al.*, *Eur. Phys. J. A* **42**, 503 (2009).
- [30] T. E. Cocolios *et al.*, *Nucl. Instrum. Methods B* **317**, 565 (2013).
- [31] T. E. Cocolios, R. P. de Groote, *et al.*, *Nucl. Instrum. Methods B* **376**, 284 (2016).
- [32] S. Rothe *et al.*, *Nucl. Instrum. Methods B* **317**, 561 (2013).
- [33] B. A. Marsh *et al.*, *Nucl. Instrum. Methods B* **317**, 550 (2013).
- [34] R. E. Rossel *et al.*, *Nucl. Instrum. Methods B* **317**, 557 (2013).
- [35] A. Coc *et al.*, *Phys. Lett. B* **163**, 66 (1985).
- [36] C. Ekström *et al.*, *Hyperfine Interact.* **4**, 165 (1978).
- [37] E. Gomez, S. Aubin, L. A. Orozco, G. D. Sprouse, E. Iskrenova-Tchoukova, and M. S. Safronova, *Phys. Rev. Lett.* **100**, 172502 (2008).
- [38] J. S. Grossman, L. A. Orozco, M. R. Pearson, J. E. Simsarian, G. D. Sprouse, and W. Z. Zhao, *Phys. Rev. Lett.* **83**, 935 (1999).
- [39] J. Zhang, M. Tandecki, R. Collister, S. Aubin, J. A. Behr, E. Gomez, G. Gwinner, L. A. Orozco, M. R. Pearson, and G. D. Sprouse, *Phys. Rev. Lett.* **115**, 042501 (2015).
- [40] M. S. Basunia, *Nucl. Data Sheets* **121**, 561 (2014).
- [41] F. G. Kondev, *Nucl. Data Sheets* **109**, 1527 (2008).
- [42] D. F. Torgerson, R. A. Gough, and R. D. Macfarlane, *Phys. Rev.* **174**, 1494 (1968).
- [43] K. Valli and E. K. Hyde, *Phys. Rev.* **176**, 1377 (1968).
- [44] D. F. Torgerson and R. D. Macfarlane, *Phys. Rev. C* **2**, 2309 (1970).
- [45] G. Neyens, *Rep. Prog. Phys.* **66**, 633 (2003).
- [46] M. D. Seliverstov, T. E. Cocolios, W. Dexters, A. N. Andreyev, S. Antalic, A. E. Barzakh, B. Bastin, J. Büscher, I. G. Darby, D. V. Fedorov, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, G. Huber, M. Huyse, M. Keupers, U. Köster, Y. Kudryavtsev, B. A. Marsh, P. L. Molkanov, R. D. Page, A. M. Sjödin, I. Stefan, P. Van Duppen, M. Venhart, and S. G. Zemlyanoy, *Phys. Rev. C* **89**, 034323 (2014).
- [47] R. Bauer, J. Speth, *et al.*, *Nucl. Phys. A* **209**, 535 (1973).
- [48] G. Ulm *et al.*, *Z. Phys. A* **325**, 247 (1986).

- [49] L. W. Wansbeek, S. Schlessner, B. K. Sahoo, A. E. L. Dieperink, C. J. G. Onderwater, and R. G. E. Timmermans, *Phys. Rev. C* **86**, 015503 (2012).
- [50] B. Cheal, T. E. Cocolios, and S. Fritzsche, *Phys. Rev. A* **86**, 042501 (2012).
- [51] F. Chappert, N. Pillet, M. Girod, and J. F. Berger, *Phys. Rev. C* **91**, 034312 (2015).
- [52] B. Bally, B. Avez, M. Bender, and P. H. Heenen, *Phys. Rev. Lett.* **113**, 162501 (2014).
- [53] J. Dobaczewski, M. V. Stoitsov, W. Nazarewicz, and P. G. Reinhard, *Phys. Rev. C* **76**, 054315 (2007).

7.1.1 Characterisations of the reference isotope, ^{221}Fr

Both low- and high-resolution measurements of ^{221}Fr are available from the 2014 IS471 experiment at the CRIS setup. While the primary objective of the experiment was to demonstrate the high-resolution available thanks to an improved laser setup [Gro15; Lyn16; Coc16], low-resolution measurements were also taken in order to compare with previous francium analyses [Fla13b; Lyn14; Bud14]. Examples of each are shown in Figure 7.4 and are both fitted differently according to the resolution type exhibited. Serving as a reminder to the statements in Chapters 4.4 and 4.4.1, high-resolution measurements have been fitted with Voigt profiles, with the angular-momentum coupling constants and the A -hyperfine parameter ratio additionally left as free variables. Low-resolution measurements, involving broadband laser light from the RILIS group, have been fitted with pure Gaussian profiles. The aforementioned parameters left as free values in the high-resolution scenario are now constrained to expected values given in Table 7.1; B -hyperfine parameters are also set to zero.

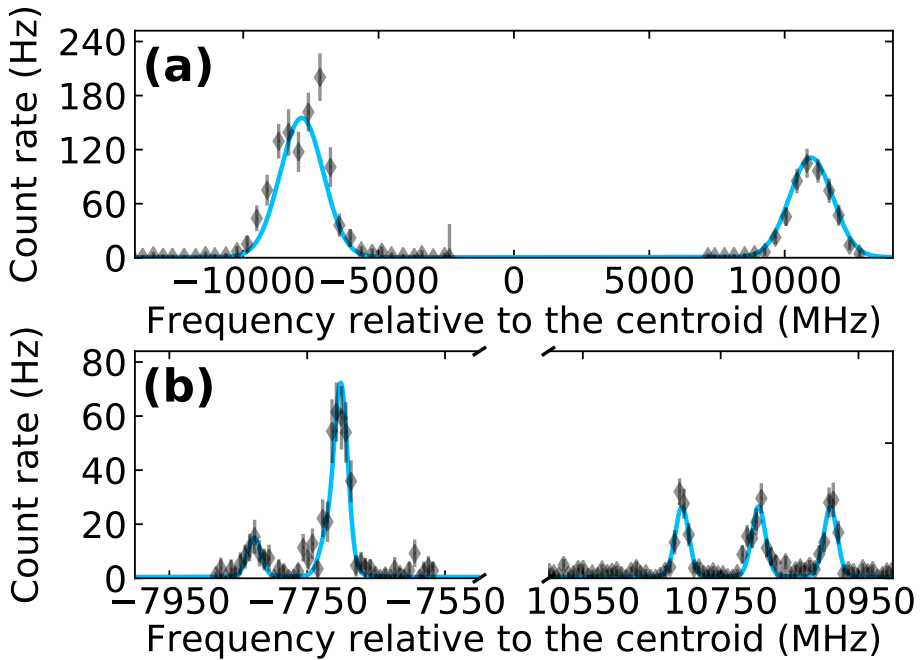


Figure 7.4: Hyperfine spectra of ^{221}Fr measured in: (a) low resolution (scans [410,411]), and (b) high resolution (scans [719,720]). Each are fitted assuming an $I = 5/2$ (blue) spin assignment.

Table 7.1: Extracted A - and B -hyperfine parameters and FWHM's for the low- and high-resolution measurements of ^{221}Fr presented in Figure 7.4. All values presented in this table are in units of MHz.

Scan Type	$A(7s\ ^2S_{1/2})$	$A(8p\ ^2P_{3/2})$	$B(8p\ ^2P_{3/2})$	FWHM
Figure 7.4(a)	+6308(33)			1871(75)
Figure 7.4(b)	+6211(1)	+22.2(3)	-84(2)	22(3)
Ref. [Gro15]	+6209(1)	+22.3(5)	-87(2)	

A summary of the A - and B -hyperfine parameters characterising each ^{221}Fr scan are given in Table 7.1. A reduction by a factor of ≥ 80 in the linewidth resolution confirms the huge strides that have been made between the francium-isotope experiments³ performed at the CRIS experiment. Considering the limited linewidth-resolution available for the measurements of ^{214}Fr in **Article II**, it would be intuitive to compare with reference measurements conducted under similar operating conditions. Unfortunately, low-resolution ^{221}Fr isotope measurements only occurred during the beginning of the experiment in which the CRIS- and laser-setup conditions had not yet been optimised, such as the ion-beam transport and laser-power stabilities for example. The $\sim 3\sigma$ deviation of its associated $A(7s\ ^2S_{1/2})$ -hyperfine parameter with respect to the tabulated value is also indicative of these experimental start-up problems before the commencement of the experiment. Based on this, the high-resolution ^{221}Fr measurement depicted in Figure 7.4(b) has been used as the reference for every ^{214}Fr hyperfine-structure measurement presented later in this thesis.

7.1.2 ^{214}Fr hyperfine scans

Four complete hyperfine-structure measurements of ^{214}Fr are presented in Figure 7.5 based on the data-file groupings declared in Table 4.2. Spectrum #1 represents the same data set published in **Article II** as Fig. 1; its selection was primarily due to its higher available statistics with respect to the others. A large variation of the hyperfine intensities and FWHM's between the observed resonances for each spectrum can be attributed to the erratic sub-optimal operating conditions experienced at the CRIS setup towards the end of the 2014 IS471 experiment. It would thus be inappropriate to analyse an amalgamated data set like for the case of ^{82}Ga in Chapter 6.3.3. Additional problems with the CRIS laser system meant that the 422.7-nm light required for the excitation step (see Figure 3.4) had to be provided instead from the RILIS laser

³Further information regarding improvements between different francium experiments can be found in Ref. [Gro17a].

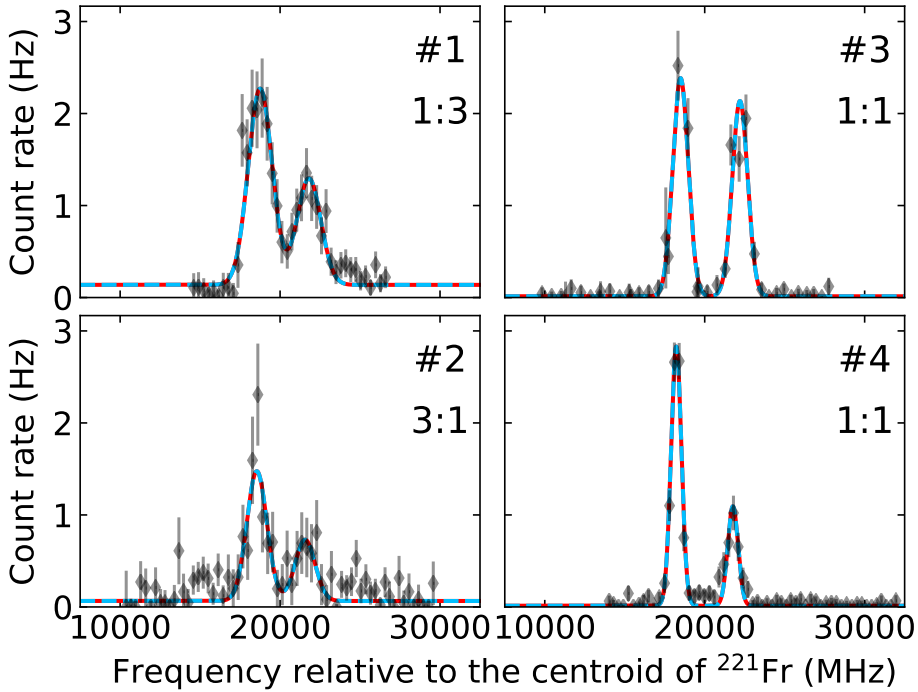


Figure 7.5: Hyperfine spectra of ^{214}Fr , each fitted assuming $I = (1)$ (red) and (2) (blue) tentative spin assignments. Aspect ratios displayed beneath each ^{214}Fr spectrum number represent the true size for each measurement. See text regarding the similar geometries for each fit of a given spectrum.

laboratory [Mar13; Rot13], explaining its rather broadband nature. Nonetheless, $A(7s\ ^2S_{1/2})$ -hyperfine parameters and isotope shifts can still be extracted, in which these values are provided in Table 7.2 for tentative $I = (1)$ and (2) spin assignments; the justification for these choices is discussed in the next section.

Table 7.2: Extracted A -hyperfine parameters and isotope shifts for ^{214}Fr , assuming tentative $I = (1)$ and (2) spin assignments. These are borne from weighted means of those extracted from the four spectra in Figure 7.5.

A	$A(7s\ ^2S_{1/2})$ (GHz)		$\delta\nu^{221,214}$ (GHz)	
	$I = (1)$	$I = (2)$	$I = (1)$	$I = (2)$
214	+2.36(9)	+1.42(5)	+19.53(9)	+19.78(10)

7.2 Nuclear-spin-assignment analysis of ^{214}Fr

Considering its location immediately after $N = 126$, ^{214}Fr serves as an ideal probe for testing the validity of shell-model principles. In particular for its ground-state spin assignment, electron-capture feeding from ^{214}Ra with $I^\pi = 0^+$ already suggests its value is low [Tor68]. This same study already excludes $I^\pi = 0^-$ because of an α transition to an excited $I^\pi = 4^+$ state in ^{210}At which would otherwise be parity forbidden. A cursory examination of the nearby $N = 127$ isotone $^{210}_{83}\text{Bi}$ reveals additional insight: an $I^\pi = 1^-$ ground state, with a long-lived low-lying $I^\pi = 9^-$ isomer state, both suggested to originate from a dominating $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration [She89]. With other $N = 127$ isotones such as $^{212}_{85}\text{At}$ and $^{216}_{89}\text{Ac}$ also suggested to exhibit similar nuclear structure with long-lived $I^\pi = (1^-)$ and (9^-) states [Lön82; Tor70], this serves as a starting point not only for the spin-assignment analysis, but also the g -factor discussion in Chapter 7.3. As further detailed in Chapters 7.2.3 and 8.2.1, spin assignments associated with the high-spin isomer, $I = (8)$ or (9) , are not considered for the analyses presented in this section.

7.2.1 Via Paar's parabola rule

Akin to Figures 6.5 and 6.6 for the cases of $^{80,82}\text{Ga}$, Figure 7.6 displays the Paar parabolas for a variety of suspected low-lying pn configurations for ^{214}Fr , based on the next predicted shell-model orbitals after a ^{208}Pb core from Figure 2.1. Additional offsets have been applied to each configuration reflecting the difference in proton and neutron SPE between the: $\pi 1h_{9/2}$ and $\pi 2f_{7/2}$ orbitals, and $\nu 2g_{9/2}$, $\nu 1i_{11/2}$, and $\nu 2g_{7/2}$ orbitals, based on Skyrme interactions which accurately reproduce the charge-radii kink at $N = 126$ for the $_{82}\text{Pb}$ and $_{84}\text{Po}$ isotope chains [God13]. Due to the negative parity which results from every pn configuration considered, all mentioned spin values are associated with this type of parity for the remainder of not only this discussion, but also for the rest of Chapter 7.

Based on the likeliest $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration, it may seem logical to conclude with an $I^\pi = 0^-$ ground state because it lies lowest in energy. However the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ and $(\pi 1h_{9/2} \otimes \nu 2g_{7/2})$ configurations are quasi-degenerate, concerning the $I^\pi = 1^-$ and $I^\pi = 8^-$ states. Their resulting repulsions from configuration mixing are likely to push the states associated with the former configuration lower in energy than the $I^\pi = 0^-$ state, thus competing with each other for the ground state. A large spin difference between these two aforementioned nuclear states also means that isomerism is likely to be present; based on the systematics seen for $^{210}_{83}\text{Bi}$, $^{212}_{85}\text{At}$, and $^{216}_{89}\text{Ac}$ as

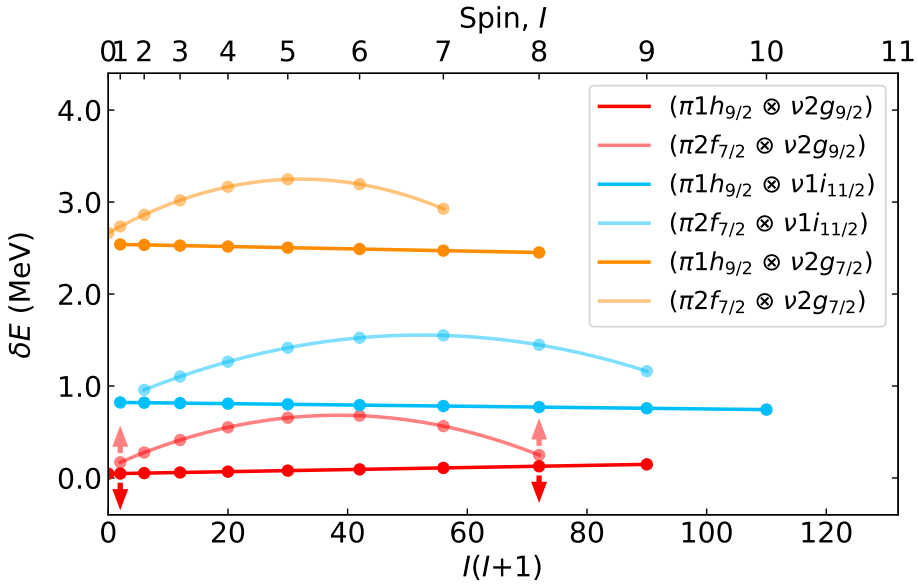


Figure 7.6: Paar parabolas for the nuclear structure of ^{214}Fr , assuming: $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ (red), $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$ (light red), $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ (blue), $(\pi 2f_{7/2} \otimes \nu 1i_{11/2})$ (light blue), $(\pi 1h_{9/2} \otimes \nu 2g_{7/2})$ (orange) and $(\pi 2f_{7/2} \otimes \nu 2g_{7/2})$ (light orange) configurations. As explained in the text, certain shell-model orbitals have been offset based on recent theoretical Skyrme-model interactions. The arrows visualise an instance of how the repulsion between alike configuration states can push certain states lower in energy; see text for further details.

described in the above introduction, this is suspected to be applicable to the $I^\pi = 8^-$ state. Overall for the nuclear structure of $^{214}_{87}\text{Fr}$, the Paar parabola methodology predicts two low-lying states with spin and parity assignments of $I^\pi = 1^-$ and 8^- .

7.2.2 Via laser-spectroscopy methods

Although a lack of high-resolution for ^{214}Fr means that no firm spin assignment analysis is possible, tentative statements can still be made involving the comparison of the observed peak intensities as detailed in Chapter 2.3.3. The inability to resolve the upper $8p \ ^2P_{3/2}$ state means that the usual relation given in (2.45) can be further simplified to:

$$\frac{S_{FF'}}{S_{JJ'}} = \frac{I+1}{I}, \quad (7.1)$$

which is solely dependent on I . Further mathematical details which allow for this simplification can be found in Appendix A.5. Two additional parameters are also introduced into the fitting function, acting as multiplicative scaling factors for each F state involved in the francium-transition scheme. Although this explains the unconventionally-equivalent lineshapes for each fitting routine for a given spectrum in Figure 7.5, regardless of the spin-assignment choice, it is (personally) seen as an easier and more direct way to extract the intensity ratio between the two observed resonances. Extracted ratios for each spectra in Figure 7.5 are displayed in Table 7.3, alongside expected ratios for $I = 1 - 12$ spin assignments, calculated from (7.1). In comparison with those predicted for the high-spin assignments, it is clear to see why distinguishing between them poses a much greater challenge.

Substantial differences between the ratios for each scan in Figure 7.5 further highlights the worsening CRIS-setup conditions experienced towards the end of the experiment, with respect to both the beamline and laser setups. This is especially prominent for scans #3 and #4, whose ratios to within 1σ do not correspond to any spin assignment. Those for scans #1 and #2 correspond to a low-spin assignment; $I = (1)$ or (2) to within 1σ . In particular for scan (1), its ratio provided the best opportunity for a tentative spin-assignment analysis in **Article II** and thus explains its presence in the aforementioned article in addition to its strongest overall statistics.

Table 7.3: Extracted intensity ratios between the two hyperfine resonances observed in the ^{214}Fr spectra displayed in Figure 7.5. Expected ratios for the $I = 1 - 12$ spin assignments using (7.1), are shown on the right.

Scan No.	Ratio	I	Ratio	I	Ratio	I	Ratio
1	1.85(28)	1	2.00	5	1.20	9	1.11
2	2.12(64)	2	1.50	6	1.17	10	1.10
3	1.12(18)	3	1.33	7	1.14	11	1.09
4	2.61(34)	4	1.25	8	1.13	12	1.08

7.2.3 Via decay-spectroscopy methods

Using the DSS1.2 setup as described in Chapter 3.3.4 and Figure 3.7, complimentary α -decay spectroscopy has also been conducted for ^{214}Fr . With a ~ 50 keV separation between the most prominent α -particle energies in both long-lived states [Ryt91], such spectroscopy is ideal for discerning the exact composition of the isotope beam for each hyperfine spectra measured in Figure 7.5. Figure 7.7 shows two separate cases: one to determine the content of the full atomic $A = 214$ beam, and another to analyse the ion beam with lasers placed on-resonance with ^{214}Fr . Each resulting spectrum represents a total of 60 minutes of collected data using the APIPS and PIPS detectors available. Laser-ionised data consists of two 30 minute measurements with the 422.7-nm laser light detuned by 18.3 GHz and 22.1 GHz respectively, in accordance with the resonance positions of scan (1) in Figure 7.5. Figure 7.7 also represents a more complete version of that shown in **Article II**.

The energy calibration of Figure 7.8 uses the most intense α transitions observed in the full-beam spectrum: ^{214}Fr , and the isobaric contaminant ^{214}Ra with its

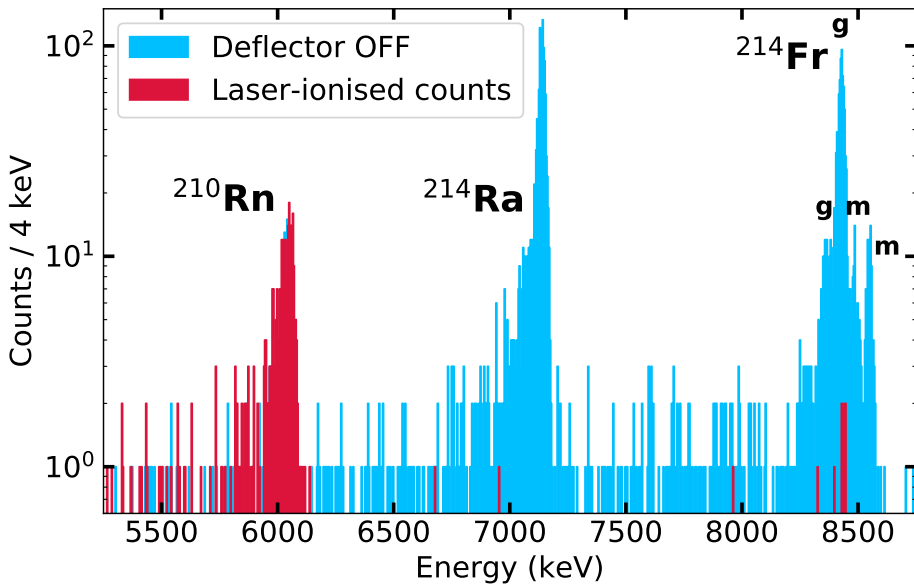


Figure 7.7: α -decay spectroscopy of the full atomic (blue) and laser-ionised (red) $A = 214$ isotope beam. Peaks are labelled according to the isotope in which the corresponding α transition belongs to; g and m identifiers distinguish between those originating from ground- and isomer-state transitions, respectively.

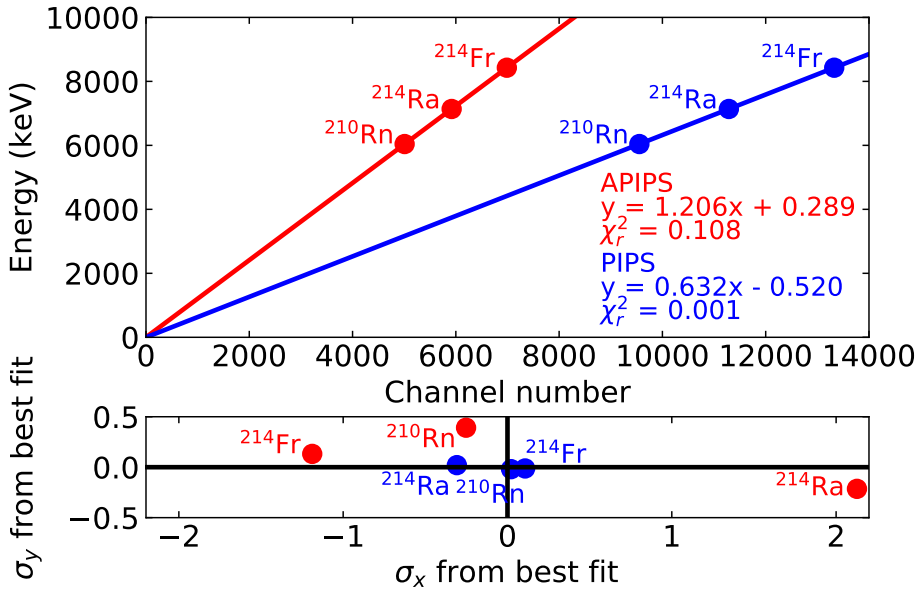


Figure 7.8: (a) Energy-calibration curves for the APIPS (red) and PIPS (blue) detectors, used for the collection of α -decay spectroscopy data presented in Figure 7.7. (b) A 2D residual plot for each data point featured above, with respect to the channel-number (x) and energy (y) uncertainties. Each data point in both subplots is labelled with its corresponding isotope.

daughter nucleus ^{210}Rn . Curves and equations for each detector calibration are shown in Figure 7.8, revealing excellent fits for the three-point calibrations⁴ using linear relationships. For the calibrated $A = 214$ spectrum, α peaks at 8365(9) keV, 8483(4) keV, and 8546(4) keV are consistent with the next strongest ^{214g}Fr (first) and ^{214m}Fr (second and third) α transitions [NND18] and adds further confidence to this calibration procedure. Although the landscape between 7250 - 8000 keV alludes to the presence of additional ^{214}Fr α transitions, they appear too weak to be conclusively included in the overall decay spectroscopy study. Despite the absence of isobaric ^{214}Ra ($t_{1/2} = 2.46(3)$ s [Bem73]) in the laser-ionised beam, the prevalence of ^{210}Rn ($t_{1/2} = 2.4(1)$ h [Cro68]) is due to the decay of impinged ^{210}Rn nuclei on the surface of the silicon detectors, from recoils during the α decay of ^{214}Ra studied from the $A = 214$ full-beam measurements beforehand.

⁴Although the 50 Bq ^{241}Am source incorporated within the DSS1.2 setup provided an additional calibration point, it is not included because of otherwise inaccurate calibrations which can be attributed to α -particle energy losses through the different copper-disk and carbon-foil substrates.

While this α -decay spectroscopy analysis confirms the unequivocal presences of both long-lived ^{214}Fr nuclear states in the full $A = 214$ beam with a ratio of 5.2(4):1 (corresponding to 84(1)% and 16(1)% respectively), only 10 ground-state α particles can be associated with the laser-ionised spectrum. Despite this, a statistical analysis can be performed to not only deduce the likelihood of this phenomena, but also for confirming the credibility of the low-value spin-assignment assumptions made in Chapter 7.2.2. Alpha-particle emission in this scenario can originate either from: the laser-ionised signal representative of one state only, or the background which consists of the same ground-/isomer-state ratio as determined for the $A = 214$ beam beforehand. For similar reasons as explained in Chapter 7.2.2, scan #1 of Figure 7.5 is chosen as a representative hyperfine spectrum with a signal and background of 6.4(7) Hz and 0.4(1) Hz respectively. Thus, the following probabilities⁵ of two scenarios can be deduced:

- The probability of a ground-state α particle, assuming a ground-state hyperfine structure: $P(\alpha_{gs}|\text{gs structure}) = 1 \times \frac{6.4}{6.8} + 0.84 \times \frac{0.4}{6.8} = 99.0^{+0.3}_{-0.4}\%$.
- The probability of a ground-state α particle, assuming an isomer-state hyperfine structure: $P(\alpha_{gs}|\text{ms structure}) = 0 \times \frac{6.4}{6.8} + 0.84 \times \frac{0.4}{6.8} = 5.1^{+1.5}_{-1.3}\%$.

A *reductio ad absurdum* methodology is first considered: supposing the hyperfine structure observed in scan #1 of Figure 7.5 is of a purely-isomeric nature, the observation of 10 α_{gs} counts analogous to that in Figure 7.7 would have an associated probability of $(5.1^{+1.5}_{-1.3}\%)^{10}$. This corresponds to a $7.4^{+0.4}_{-0.3}\sigma$ significance event; a timeframe of at least 22 million years might be needed to detect such an occurrence. Based on this, observing pure isomer-state structures for the spectra shown in Figure 7.5 can be ruled out with a near-100% confidence. Although the possibility of a mixed-state beam cannot be ignored *a priori*, a factor of ~ 5 suppression of the isomer state from the α -decay analysis of the $A = 214$ beam means that any corresponding hyperfine structure can be neglected as it would be comparable with the background from Figure 7.5. Following this, a *lex parsimoniae*⁶ methodology is additionally considered: the probability of 10 α_{gs} counts corresponds to simply $P(\alpha_{gs}|\text{gs structure})^{10} = 90.6^{+3.1}_{-3.9}\%$. Overall, the favourable likelihood of observing a low-spin ground-state structure reinforces the use of $I = (1)$ and (2) spin assignments in Chapter 7.2.2 and for the subsequent analyses regarding both the magnetic-dipole moment and the change in its mean-square charge radii.

⁵Considering only α_{gs} and α_{ms} are possible, it is therefore trivial that $P(\alpha_{ms}|\text{*s structure}) = 1 - P(\alpha_{gs}|\text{*s structure})$.

⁶More commonly known as Occam's razor.

7.3 g factor of ^{214}Fr

It is first reminded that a full electromagnetic-moment analysis for ^{214}Fr only extends to the g factor because the lack of sensitivity to the $B(8p\ ^2P_{3/2})$ -hyperfine parameter prevents the extraction of an electric-quadrupole moment.

Article II details the initial g -factor analysis based on the comparison with empirical g factors calculated from (2.17), involving the likeliest proton and neutron shell-model orbitals after $Z = 82$ and $N = 126$: $\pi 1h_{9/2}$ and $\nu 2g_{9/2}$, respectively. Configuration mixing arising from the $\nu 1i_{11/2}$ orbital had also been considered because of its close proximity to the $\nu 2g_{9/2}$ orbital, based on theoretical studies of relevant $_{82}\text{Pb}$ and $_{84}\text{Po}$ isotopes [God13].

In this section however, an updated g -factor analysis now extends the model space to incorporate the $\pi 2f_{7/2}$ and $\nu 2g_{7/2}$ shell-model orbitals; this amalgamation is pictorially described in Figure 7.9. While the latter has been included for completeness due to investigations of its involvement as an admixture-contribution term (see Appendix A.4), the addition of the $\pi 2f_{7/2}$ orbital is motivated by a recent theoretical survey of the ^{208}Pb region with LSSM

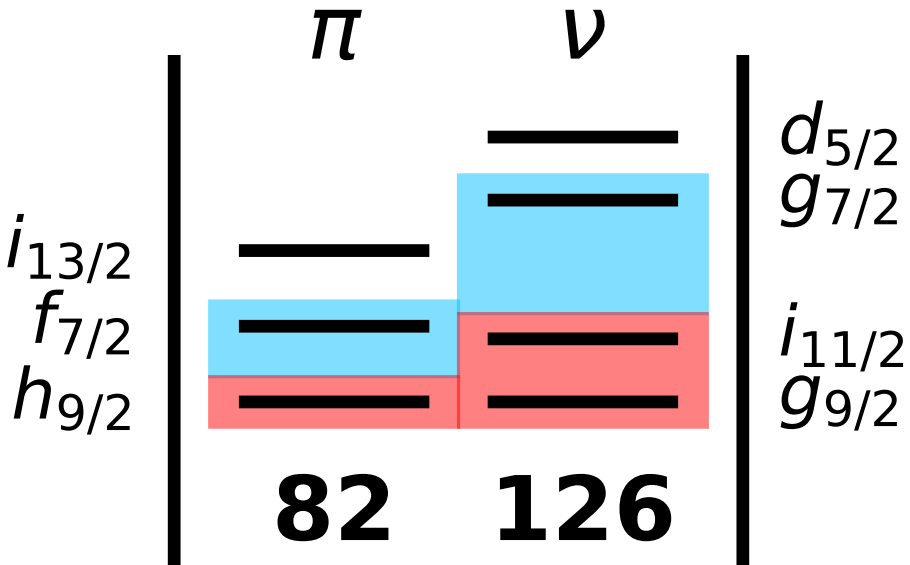


Figure 7.9: An overview of the shell-model orbitals which feature past $Z = 82$ and $N = 126$. The red- and blue-shaded areas represent the model spaces considered for the g -factor analyses in **Article II** and this chapter, respectively.

calculations, detailed in Ref. [Yan18a]. Intriguingly for ^{214g}Fr assuming an $I = 1$ spin assignment, the aforementioned reference proposes a $(\pi 1h_{9/2}^4 2f_{7/2} \otimes \nu 2g_{9/2})$ ground-state configuration⁷, despite its theoretically-predicted g factor matching comparably with the experimental value reported in **Article II**, agreeing favourably with a $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration. Even though these recent developments had been published after **Article II**, updating existing interpretations to account for the on-going study of a particular topic should be designated as a matter of utmost importance (see Chapter 7.4 as another example). Single-particle magnetic-dipole moments for the involved shell-model orbitals are catalogued in Table 7.4, while the numerical and graphical summaries of the ^{214}Fr g -factor analysis are given in Table 7.5 and Figure 7.10 respectively. These compare experimental and (available) LSSM values for the tentative $I = (1)$ and (2) spin assignments, with empirical calculations for various pn configurations. The latter infographic in particular, serves as a direct extension to Fig. 4 in **Article II**.

Regardless of the tentative spin-assignment choice, the ground state of ^{214}Fr can be predominately described by a $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration, akin to other nuclear states within the francium-isotope multiplet involving the

Table 7.4: Representative single-particle magnetic-dipole moments for the shell-model orbitals featuring in the extended model space from Figure 7.9. No uncertainties are given for the theoretical values, *id est* in Ref. [Bau73].

Orbital	μ (μ_N)	Isotope	Ref.
$\pi 1h_{9/2}$	+4.02(1)	$^{213}_{87}\text{Fr}_{126}$	[Coc85]
$\pi 2f_{7/2}$	+5.338 ⁸	$^{209}_{83}\text{Bi}_{126}$	[Bau73]
$\nu 2g_{9/2}$	-1.197(85)	$^{211}_{84}\text{Po}_{127}$	[Sel14]
$\nu 1i_{11/2}$	+0.695 ⁸	$^{209}_{82}\text{Pb}_{127}$	[Bau73]
$\nu 2g_{7/2}$	+0.945 ⁸	$^{209}_{82}\text{Pb}_{127}$	[Bau73]

⁷Private correspondence with the author of Ref. [Yan18a], Kota Yanase, revealed that wavefunction amplitudes cannot be directly calculated by their computational codes. He mentioned that configurations are instead inferred by the orbital occupation numbers which for $^{214}\text{Fr}(I^\pi = 1^-)$:

- $\pi 1h_{9/2}$: 3.86, $\pi 2f_{7/2}$: 0.57, $\pi 1i_{13/2}$: 0.44, $\pi 2f_{5/2}$: 0.08, $\pi 3p_{3/2}$: 0.04, $\pi 3p_{1/2}$: 0.02.
- $\nu 2g_{9/2}$: 0.96, $\nu 1i_{11/2}$: 0.00, $\nu 1j_{15/2}$: 0.00, $\nu 3d_{5/2}$: 0.04, $\nu 4s_{1/2}$: 0.00, $\nu 2g_{7/2}$: 0.00.

⁸For comparison of Schmidt estimates from (2.16) and using $g_{s,eff} = 0.7g_{s,free}$ as suggested in Ref. [Ney03]: $\mu_{Schmidt}(\pi 2f_{7/2}) = +4.955\mu_N$, $\mu_{Schmidt}(\nu 1i_{11/2}) = +1.133\mu_N$, and $\mu_{Schmidt}(\nu 2g_{7/2}) = +1.042\mu_N$. The Bauer moments from Ref. [Bau73] nonetheless, have been used because they are assumed to more representative of any quenching effects that are local to the ^{208}Pb region.

Table 7.5: Extracted g factors for ^{214}gFr assuming tentative $I = (1)$ and (2) spin assignments, using the A -hyperfine parameters catalogued in Table 7.2 (first line). Where applicable, these are compared with: LSSM calculations from Ref. [Yan18a] (second line), and various pn configurations (from the third line onwards) calculated from values quoted in Table 7.4.

Method	g factor	
	$I = (1)$	$I = (2)$
Expt.	+0.239(10)	+0.144(6)
LSSM	+0.232	
$(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$	+0.314(10)	+0.314(10)
$(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$	-1.599(6)	-0.193(2)
$(\pi 1h_{9/2} \otimes \nu 2g_{7/2})$	+1.984(5)	+1.049(2)
$(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$	-3.401(44)	-0.714(17)

$\nu 2g_{9/2}$ orbital⁹. This also suggests that the ground state has a negative parity. Additional contributions for $I^\pi = (1^-)$ and (2^-) can arise from either: a $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ contribution requiring 96%/4% and 67%/34%, or a $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$ configuration requiring 98%/2% and 83%/17%, respectively. Considering the remarkable purity of the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration resulting from the $I^\pi = (1^-)$ options, along with this expectation within the realms of shell-model principles near suspected shell closures, an $I^\pi = (1^-)$ spin assignment is thus suggested for ^{214}gFr . This also matches with predictions based on the Paar parabola methodology in Chapter 7.2.1. Based on the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ g -factor positions, this configuration cannot contribute to the total g factor for both spin assignments assuming this analysis; see Appendix A.4 for further insight into how this configuration could contribute to the g factor.

Discerning the exact composition of the additional contributions required to completely explain the $I^\pi = (1^-)$ g factor however, remains ambiguous. Those originating from $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ and $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$ configurations are both plausible, considering that only a few % is needed from either one. While the definitive $I^\pi = 9/2^-$ ground-state spin assignments for the neighbouring ^{213}Fr and ^{215}Fr isotopes [Eks78; Sch84] adds credence to the former configuration, the results of the shell-model studies in Ref. [Yan18a] cannot be ignored. A $(\pi 1h_{9/2}^4 \pi 2f_{7/2})_{I^\pi=9/2^-}$ proton configuration of course, could arise from pair-breaking within the $\pi 1h_{9/2}$ orbital. However it is surprising that the

⁹Although a magnetic-dipole moment is known for ^{220}Fr , its $I^\pi = 1^+$ spin-parity assignment cannot be explained by the suggested dominating configuration mentioned in Figure 7.10. It belongs to the parity-doublet band $K^\pi = 1^+$, indicative of a region with stable octupole deformations [She88; Lia92].

negative $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})_{1-}$ g -factor value in Table 7.4 and Figure 7.10 directly contradicts the theoretically-calculated positive value in Ref. [Yan18a]. At present, this remains an open question. A high-resolution measurement of ^{214g}Fr , yielding a firm spin assignment and an electric-quadrupole moment, would help to fully characterise its nuclear structure. Additional spectroscopy measurements and theoretical studies would help to not only discern the SPE of the orbitals

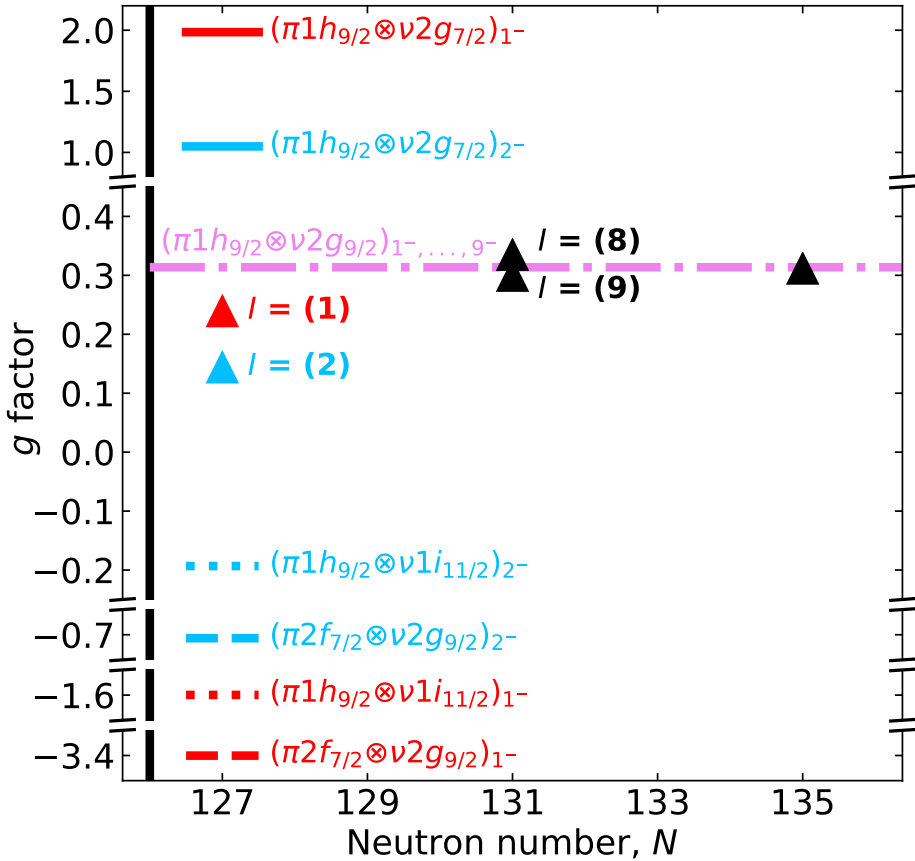


Figure 7.10: Extracted ^{214g}Fr g factors for $I = (1)$ (red) and (2) (blue) assignments, alongside those for other francium isotopes (black) with semi-filled $\nu 2g_{9/2}$ shells (^{218m}Fr : Ref. [Bud14], ^{222}Fr : Ref. [Coc85]). These are compared with empirical values assuming pure: $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ (dash-dotted line), $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ (dotted lines), $(\pi 1h_{9/2} \otimes \nu 2g_{7/2})$ (solid lines), and $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$ (dashed lines) configurations, for the applicable spin assignments.

involved in this study, but also confirm which are pivotal for explaining the g factor. Further intricacies regarding the neutron-orbital contributions towards the total g factor is discussed in Appendix A.4.

7.4 Change in the mean-square charge radii of ^{214}Fr

Changes in the mean-square charge radii for ^{214g}Fr are displayed in Table 7.6, for the $I = (1)$ and (2) spin assignments. Its inclusion in Figure 7.3 revealed a characteristic kink immediately after $N = 126$, analogous to the behaviours of other isotope chains near several shell closures throughout the nuclear landscape [Cam16]. A revision of the F and M atomic parameters originally reported in Ref. [Lyn14] for the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition explains the differences between values reported in Table 7.6 with those in **Article II**. Further details, as well as a comprehensive evaluation of changes in the mean-square charge radii for all francium isotopes measured at the CRIS setup, can be found in Appendix A.6.

Since the publication of **Article II**, additional charge radii measurements have been made in the near vicinity of $N = 126$. From Table 7.7, new data points are now available for ^{80}Hg and ^{85}At ; those marked with an asterisk (*) originate from Ph.D theses (Refs. [Tei17; Day17]) and thus, represent isotope-shift measurements which are yet to be published and are not the finalised values. Nonetheless, this still highlights a continuing interest of characterising the $N = 126$ shell closure. Given the large statistical uncertainties that normally bestow measurements conducted in low-resolution, the occurrence of multiple measurements for a particular isotope must also not be ignored. Where applicable, values in Table 7.7 represent a weighted mean of: either the isotope shifts (for measurements on the same transition¹⁰: ^{80}Hg , and ^{85}At), or

Table 7.6: Changes in the mean-square charge radii for ^{214}Fr , assuming the tentative $I = (1)$ and (2) spin assignments. These have been calculated using the extracted isotope shifts in Table 7.2, along with the F_{422} and M_{422} atomic parameters given in Appendix A.6.

I	$\delta\langle r^2 \rangle^{221,214} \text{ (fm}^2\text{)}$
(1)	$-0.951(4)\{10\}$
(2)	$-0.964(5)\{10\}$

¹⁰Subsequent changes in the mean-square charge radii have been calculated using the most recent F and M atomic parameters available for a given element.

Table 7.7: Additional data for changes in the mean-square charge radii immediately before and after the $N = 126$ shell closure, obtained since the publication of **Article II**. It complements and where applicable, supersedes tabulated data from Table II of the aforementioned article. See text for further details regarding the obtaining of values marked with an asterisk (*).

Element	$\delta\langle r^2 \rangle^{125,126}$ (fm ²)	$\delta\langle r^2 \rangle^{126,127}$ (fm ²)	Refs.
Hg	0.0654(34)	0.1012(53)*	[Ulm86; Day17]
Po	0.0813(10)	0.1040(100)	[Kow91; Sel13]
At	0.0772(8)	0.1300(300)*	[Tei17; Cub18]
Fr	0.0788(2)	0.1041(45)	[Coc85; Duo87] + This thesis

the charge radii themselves (for measurements involving different transitions: ⁸⁴Po, and ⁸⁷Fr). An incorrect interpretation of the statistical uncertainties made in **Article II** for ⁸⁴Po has also been rectified. Overall, Figure 7.11(a) represents an updated version of Fig. 5 in **Article II** which incorporates these new/updated values.

Although this particular charge-radii characterisation has garnered interest elsewhere, different formalisms have been suggested. The Ph.D thesis of T. Day Goodacre in Ref. [Day17], and the study in Barzakh *et al.* Ref. [Bar18] for example, argue that properly assessing the shell effect necessitates the removal of any OES influences. This involves examining instead, the ratios of changes in the mean-square charge radii between $N = 128,126$ with $N = 126,124$ (ξ_{even}), and $N = 127,126$ with $N = 125,124$ (ξ_{odd}). These parameters are defined by:

$$\xi_{even} = \frac{\delta\langle r^2 \rangle^{128,126}}{\delta\langle r^2 \rangle^{126,124}}, \quad \xi_{odd} = \frac{\delta\langle r^2 \rangle^{127,126}}{\delta\langle r^2 \rangle^{125,124}}. \tag{7.2}$$

Table 7.8: Available ξ_{even} and ξ_{odd} parameters calculated from (2.10), for elements in the vicinity of $N = 126$. See text for further details regarding their definition, or Ref. [Bar18]. Charge-radii data used their calculation have been taken from references catalogued in Appendix A.3.

Hg	Tl	Pb	Bi	Po	At	Fr, $I =$	
						(1)	(2)
ξ_{even}	2.22(9)	1.79(2)	1.86(5)				
ξ_{odd}	2.93(23)	1.97(48)	2.06(6)	2.06(14)	2.30(27)	2.36(69)	2.44(11) 2.15(11)

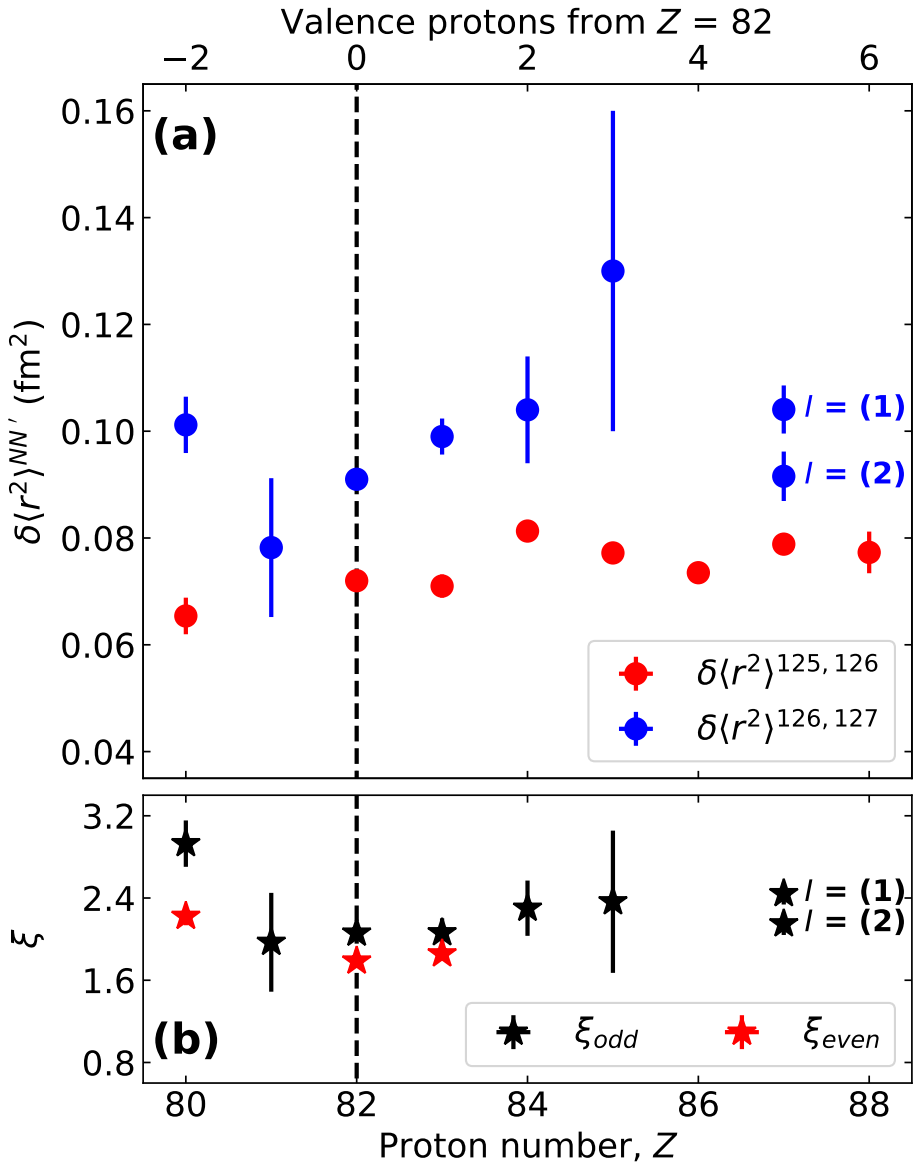


Figure 7.11: (a) Changes in the mean-square charge radii for before (red circles) and after (blue circles) the $N = 126$ shell closure, across the $N = 125$ and 127 isotone chains respectively. (b) Shell-effect parameters ξ_{odd} and ξ_{even} (black and red stars), for the same isotopes involved in the above subfigure; see text for their definitions. Only statistical uncertainties are shown in both subfigures.

While in hindsight the analysis made in **Article II** did not attempt to address this, the purpose of Fig. 5 (see Figure 7.11) was to try and better illustrate the kink at $N = 126$ than in Figure 7.3. Nonetheless for completeness, these values are numerically and graphically displayed in Table 7.8 and Figure 7.11(b) respectively; together they serve as an update to their respective counterparts that feature in Ref. [Bar18].

In general for Figure 7.11(a), $\delta\langle r^2 \rangle^{126,127}$ values are larger than $\delta\langle r^2 \rangle^{125,126}$ values for applicable isotopes; this characteristic ‘kink’ [Cam16] reinforces the magical expectations for the $N = 126$ shell closure. Theoretically, the mechanism of this kink could be successfully replicated in isotope chains using Skyrme forces with an $\nu 1i_{11/2}$ orbital occupation of at least 3% [God13]. Coincidentally in Chapter 7.3, a similar occupation can be used to explain the g factor for ^{214g}Fr , assuming the preferred spin assignment $I = (1)$. However comparing Figure 7.11(a) with Fig. 5 of **Article II**, no trend lines are now provided because the inclusion of additional data provided by Table 7.7 casts doubt on a purported Z -dependence of this kink after $N = 126$. This is further claimed in Ref. [Bar18], which found no distinction between the ξ parameters defined in (7.2) and suggesting an indiscriminate shell effect for isotopes at ($N = 126$) and after ($N = 127$) the shell closure. Despite a matching trend between ξ_{even} parameters with theoretical calculations in Fig. 3 of the aforementioned reference, a thorough comparison is still limited due to the scarcity of ξ_{even} parameters; with conventional laser-spectroscopy techniques, the ns-lived $N = 128$ isotones past $_{83}\text{Bi}$ are wholly inaccessible. A replication of this study in Figure 7.11(b) not only found statistically significant differences between the ξ_{even} and ξ_{odd} values for $_{80}\text{Hg}$ and $_{82}\text{Pb}$, but also hints at a slight Z -dependence of the ξ_{odd} parameters. The new addition of $_{80}\text{Hg}$ data also highlights an unexpected increase in its ξ_{odd} parameter compared with the rest of the landscape and could be indicative of an observable shell effect at $Z = 82$. Nonetheless without further uncertainty improvements, it is rather difficult to make firm conclusions and highlights the imperative need for high-resolution isotope measurements for a more complete characterisation.

7.5 Summary

At first glance, the study of just one isotope may initially seem insubstantial with respect to a full isotope chain synonymous with a typical laser-spectroscopy study. The study of ^{214}Fr however, represents a milestone as the shortest-lived isotope to have been measured with such techniques with a half-life of only 5 ms. It has provided a surprising wealth of information relating to the

magical extent of ^{208}Pb , despite a large linewidth resolution preventing either an electric-quadrupole moment or firm spin assignment from being measured.

Bolstered with complimentary α -decay spectroscopy measurements and Paar parabola analysis, the structure observed in spectra for ^{214}Fr likely belongs to the ground-state with a spin assignment of either $I = (1)$ or (2) . A subsequent g -factor analysis suggests a dominating $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration with a negative parity: $> 96\%$ and $> 66\%$ respectively for the aforementioned spin assignments. Assuming the former spin assignment, its pure nature is remarkable given the substantial filling of the $\pi 1h_{9/2}$ orbital past $Z = 82$; nonetheless its robustness reinforces the prestige of shell model principles. The $(\pi 1h_{9/2} \otimes \nu 1i_{11/2})$ and $(\pi 2f_{7/2} \otimes \nu 2g_{9/2})$ configurations can also contribute to the g factor for both spin assignments, however their exact influences remain unknown despite additional theoretical insights which are now available. Remeasuring this isotope with a higher resolution could help to further pinpoint this nuclear structure problem: not only with an electric-quadrupole moment measurement and a firm spin assignment, but it could also reveal the high-spin isomer which was not observed in these studies.

With respect to its change in the mean-square charge radii, ^{214}Fr represents the heaviest $N = 127$ to exhibit a kink in the values past $N = 126$, albeit reduced for $I = (2)$. While additional measurements and interpretations have since added further discussion to the charge-radii picture immediately after $N = 126$, large statistical uncertainties and missing measurements still stifle the debate regarding a possible Z dependence on the magnitude of the kink. Further measurements are needed for conclusive statements to be made.

Chapter 8

On the path to further understanding nuclear structure

Although the physics discussions presented in Chapters 5, 6, and 7 for $_{31}\text{Ga}$ and $_{87}\text{Fr}$ isotopes have contributed in the concerted efforts to characterise regions near doubly-magic cores, by no means do they represent closure for their investigations. On the contrary, other exploratory avenues remain which could help to fully explain the nuclear structure questions that would otherwise persist. With respect to the already presented analyses of $_{31}\text{Ga}$ and $_{87}\text{Fr}$ isotopes, this chapter will elaborate on the recommendations made in the ending summaries for the aforementioned chapters. Further attempted measurements, discussions, calculations, and simulations will not only highlight their necessities, but will also crucially assess the current feasibility of the CRIS setup.

N.B. All of the simulated hyperfine-spectroscopy measurements presented in this chapter are representative of the current capability of the CRIS experiment at the ISOLDE facility. Unless explicitly stated or further detailed upon, the following assumptions have been made for every simulation:

- Averaged throughout the duration of a typical experiment, a $1.5\ \mu\text{C}$ proton-beam current can be delivered by the PSB. The transmission of an isotope beam through the ISCOOL is 70%.
- A 1% total CRIS experimental efficiency and a 10^7 collisional-background suppression in the IR, can be achieved based on Refs. [Gro17b; Kos19a].

- A - and B -hyperfine parameters for a particular isotope have been calculated from sensible electromagnetic-moment estimates using (2.40) and (2.42) respectively, along with a suitable reference isotope. These latter estimates are given in the text.

8.1 For gallium isotopes

Simulated gallium-isotope measurements in this section initially assumes a CRIS-setup efficiency similar to that obtained during the first half of the 2015 IS571 experiment in Chapter 4.1.1, $\sim 1/1000$, but additionally factoring in the potential improvement in the neutralisation efficiency by switching from potassium to sodium alkali vapour as mentioned in Chapter 3.3.1: a total efficiency of $\sim 1/400$. This gain also corresponds with gallium atomic-population simulations assuming potassium and sodium as the alkali-vapour choices, in Ref. [Ver19a]; the aforementioned reference also catalogues the findings for the majority of elements in the periodic table. Exhibited lineshapes have also been fitted with Voigt profiles whose individual Gaussian and Lorentzian contributions represent purported values that can be achieved for future gallium experiments at CRIS. These are: a 20 MHz Gaussian contribution based on the resolution obtained during recent francium experiments that utilised similarly produced 422.7-nm light [Wil17a], and a 35 MHz Lorentzian contribution borne from the natural linewidth of the excited $5s\ ^2S_{1/2}$ state with a $t_{1/2} = 5.7(6)$ ns lifetime [Saf06], plus an additional contribution to account for potential power-broadening effects.

8.1.1 $^{61,62}\text{Ga}$: Towards the vicinity of ^{56}Ni

Despite the unbound nature for ^{59}Ga (see Chapter 5.3), obtaining the electromagnetic moments for ^{61}Ga would still allow the magicity of the ^{56}Ni region to be probed in conjunction with their $_{29}\text{Cu}$ isotone counterparts. Alongside ^{62}Ga , their changes in the mean-square charge radii would be beneficial for not only inferring the characteristic trends typically observed near magic shell closures [Cam16], but also investigating a possible proton-skin formation for the most neutron-deficient species [Lép05; Pro12]. With $N = Z$ status, a measurement of ^{62}Ga could additionally help to further constrain the unitarity of the Cabibbo-Kobayashi-Maskawa matrix used in the Standard Model of particle physics, by improving on the up-down element V_{ud} uncertainty [Man11b; Har15].

Although measurements on ^{62}Ga had been attempted during the 2017 IS571 experiment (see Table 4.1), the combination of ~ 8 hours worth of data in

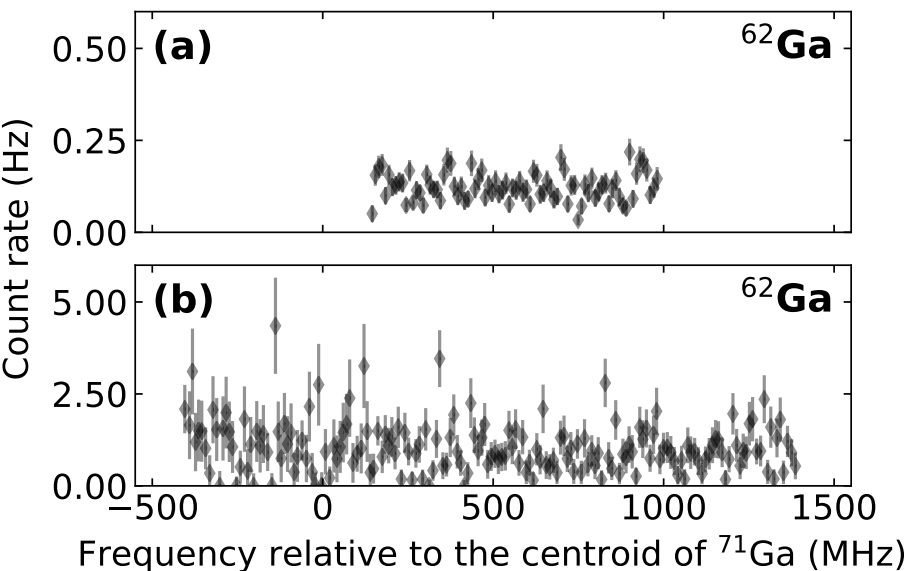


Figure 8.1: (a) The combined spectrum for the attempted hyperfine-structure measurement of ^{62}Ga . (b) A single ^{62}Ga measurement representing the largest span in frequency space.

Figure 8.1 fails to show any indications of discernible hyperfine structure. The background observed in these scans is likely to comprise of molecular contaminants such as TiO^+ and CaF^+ , similar to the cases of $^{65,67}\text{Ga}$ in Figure 5.4; based on recent ISOLTRAP mass measurements of ^{24}Cr isotopes, these impurities are produced with a $\text{TiO}^+/\text{CaF}^+ \sim 20$ ratio at $A = 63$ [Mou18]. Besides, it is much more preferable to use metal-foil targets over UC_x for producing neutron-deficient species. In particular, ZrO_2 material has already been utilised to produce $^{61,62}\text{Ga}$ isotope beams at the ISOLDE facility [Wei02;

Table 8.1: Expected $^{61,62}\text{Ga}$ yields from Ref. [Kös03] using ZrO_2 target material, alongside background-limit estimates.

A	Ga yield (μC^{-1})	FC reading before the ISCOOL (pA)
61	1.0×10^1	0.3 ¹
62	4.0×10^3	1.2

¹This represents an estimated value of $\sim 5/21 \times 1.2$ pA. Two assumptions are made: the absence of a stable $A = 45$ titanium isotope coupled with the $\text{TiO}^+/\text{CaF}^+$ ratio mentioned

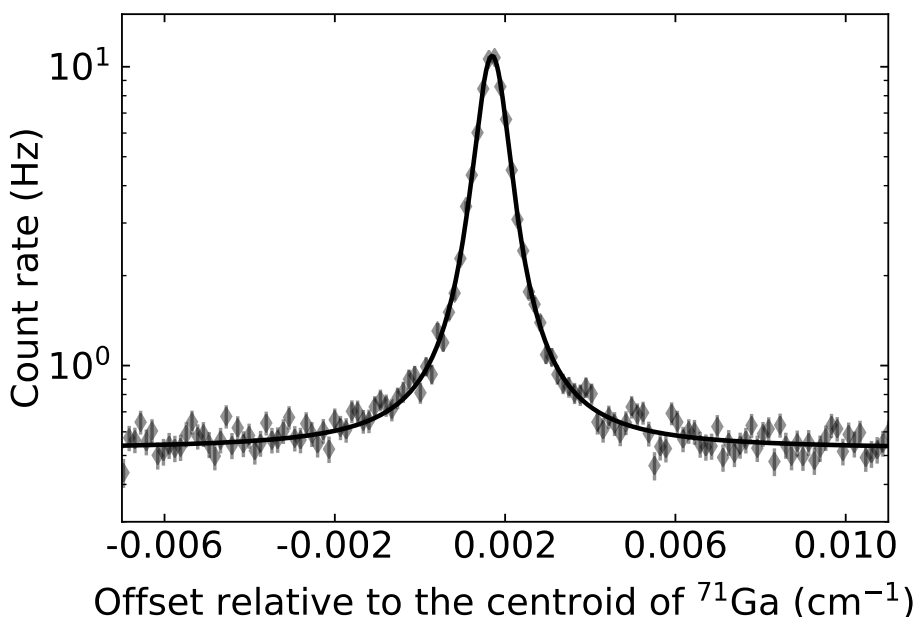


Figure 8.2: A simulated spectrum of the hyperfine structure of ^{62}Ga , fitted assuming an $I = 0$ (black) spin assignment. The estimated time required to experimentally replicate this simulation spanning 0.018 cm^{-1} with 135 steps is ~ 7 hours.

Kös03]; typically-produced yields, which are documented in Table 8.1, have been utilised for the hyperfine-structure simulations of these isotopes. FC readings taken before the ISCOOL are also given in which background-level estimations can be made. Although these quantities are associated with a UC_x target and are thus not directly comparable to those from a ZrO_2 target, they currently represent the best estimates until further characterisations are made with the latter material. The contamination composition is assumed to arise from the same molecular compounds mentioned for the UC_x targets; while enhanced isobaric copper and zinc yields towards the $\sim 10^8\text{ }\mu\text{C}^{-1}$ limit are associated with ZrO_2 targets [Kös00a; Kös03], their higher ionisation potentials prevent them from surface ionising easily.

Figures 8.2 and 8.3 display simulated spectra for ^{62}Ga and ^{61}Ga respectively, using the numbers in Table 8.1 and the assumptions made earlier in this section. While no hyperfine parameter predictions are needed for ^{62}Ga with $I = 0$,

in the text, and a natural-abundance ratio of $^{42}\text{Ca}/^{43}\text{Ca} \sim 5$ for the stable CaF^+ compounds involved at $A = 61$ and $A = 62$.

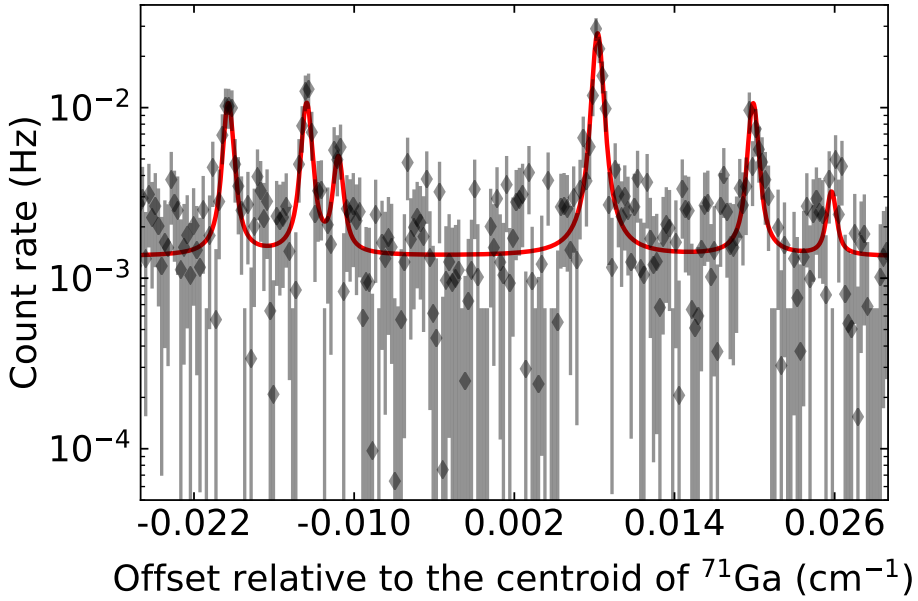


Figure 8.3: A simulated spectrum of the hyperfine structure of ^{61}Ga , fitted assuming an $I = 3/2$ (red) spin assignment. The estimated time required to experimentally replicate this simulation spanning 0.056 cm^{-1} with 235 steps is ~ 98 hours.

electromagnetic-moment values² of $\mu = +2.23 \mu_N$ and $Q_s = +19.4 \text{ efm}^2$ have been assumed for the structure of ^{61}Ga with $I = 3/2$. Isotope shifts for both isotopes assume a continued OES similar to that observed for $^{63,64,65}\text{Ga}$ in Refs. [Pro12; Far17], however considering the large deviation in the isotope shift for $N = Z$ $^{74}_{37}\text{Rb}$ with respect to the local rubidium trend [Man11b], caution should be exercised regarding similar manifestations that could be present for $^{62}_{31}\text{Ga}$. Nonetheless, a relatively precise ^{62}Ga measurement as depicted in Figure 8.2 can be obtained within the confines of a single ISOLDE shift³; even with an unpredictable isotope shift (thus enlarging the frequency space for scanning), the increased time needed would still be deemed acceptable for a typical CRIS experiment. However for measuring ^{61}Ga , almost twelve shifts would be required to replicate a spectrum with a similar data quality as in Figure 8.3, provided the background suppression is additionally enhanced by

²For μ : using the prediction in Ref. [Wei02]. For Q_s : based on the ^{63}Ga value in Ref. [Pro12], with an added deviation such that the difference between $^{61,63}\text{Ga}$ is the mirror opposite of the neighbouring isobars $^{59,61}\text{Cu}$ in Ref. [Vin11b].

³One ISOLDE shift is equivalent to 8 hours.

a factor of 100: overall, to the $\sim 10^9$ level. Thus at present, measuring ^{61}Ga would be considered untenable within the operation realms of a standard CRIS experiment. Alternatively, $^{61,62}\text{Ga}$ are ideal candidates for β -decay assisted laser-spectroscopy measurements; background levels could potentially be negligible because neither stable-molecular contaminants nor any nearby isobars with high ionisation potentials would be able to contribute. Similar concepts have already been utilised to measure ^{52}K [Kos19a] and it is strongly recommended to consider the feasibility of this technique for any future neutron-deficient gallium measurements.

8.1.2 $^{83,84}\text{Ga}$: Exploring further past $N = 50$

Seldom ground-state properties are available for isotopes residing in the region past $N = 50$ near ^{78}Ni , with ^{82}Ga as the only exception [Che12a]. Venturing deeper into this territory is required for answering the long-standing nuclear-structure questions that still remain for the gallium-isotope chain. While measuring the hyperfine structure of ^{83}Ga would test the persistence of an $I = 5/2$ ground-state akin to the situation between $^{73,75,77}\text{Cu}$ [Fla13b; Gro17b], additionally extending to ^{84}Ga could also confirm the first experimental instance of long-lived isomerism in the area [Leb09].

Measurement attempts for ^{83}Ga as summarised in Table 4.1, are displayed in Figure 8.4; the count-rate difference is attributed to a proton-beam-intensity reduction from the PSB, in-between data collections. Larger background-count rates compared to those observed for the preceding $^{81,82}\text{Ga}$ isotopes in Figures 6.2 and 6.11 highlight the increasing presences of isobaric ^{37}Rb and ^{38}Sr contaminants, which are known to be prevalent at these mass regimes. The choice of a coarse frequency-bin size for these measurements during the 2015 IS571 experiment is also unfortunate. In particular for Figure 8.4(b), the possible hyperfine structure observed could be interpreted as the, albeit rather weak, presence of ^{83}Ga . However the additional time needed (with a finer frequency-bin size) for further clarification would most likely have exceeded the length of that CRIS-setup experiment, thus compromising the measurements of $^{65,67}\text{Ga}$ which were published in **Article I**.

Figures 8.5 and 8.6 display simulated spectrums for ^{83}Ga and ^{84}Ga respectively, using the yield and contamination numbers for these mass regimes in Table

⁴Estimate values are used for ^{84}Ga . Considering the yield measurement for ^{83}Ga from the 2015 IS571 experiment (see Chapter 4.1.1) matches with the upper limit catalogued in the ISOLDE yield database [Bal19], the same assumption has been made for ^{84}Ga from the aforementioned reference. With the same database also reporting similar yield intensities for ^{37}Rb and ^{38}Sr at $A = 83$ and 84 , a similar 1.0 pA FC reading for ^{84}Ga is assumed, however it is stressed that this should be treated as a conservative estimate.

Table 8.2: Expected $^{83,84}\text{Ga}$ yields based on target-team measurements at the ISOLDE facility (see Chapter 4.1.1) using UC_x target material, alongside background-limit estimates.

A	Ga Yield (μC^{-1})	FC reading before the ISCOOL (pA)
83	6.0×10^3	1.0
84^4	8.0×10^1	1.0

8.2. Unlike the case for ^{61}Ga in Chapter 8.1.1, no electromagnetic-moment estimates are available for gallium isotopes beyond ^{82}Ga . Extrapolating from the even- and odd- A trends would be unwise because of the unknown changes in the ground-state properties that would occur after crossing $N = 50$. Therefore, assuming similar nuclear structures to the nearest available gallium isotopes is seen as the most sensible, and only, approximation to make: ^{83}Ga assumes a similar $I = 5/2$ ^{81}Ga structure, while ^{84m}Ga assumes a similar $I = 3$ ^{80m}Ga structure. Akin to the situation assumed for ^{62}Ga in Chapter 8.1.1, no hyperfine-

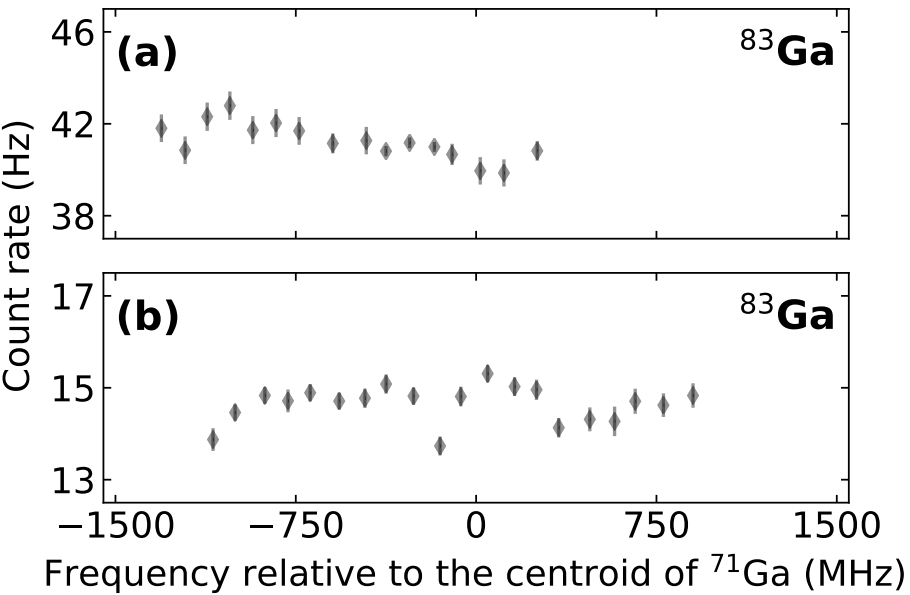


Figure 8.4: Combined spectra for attempted hyperfine-structure measurements of ^{83}Ga , using from Table 4.1: (a) scan numbers [56,57,58], and (b) scan numbers [59,60,61].

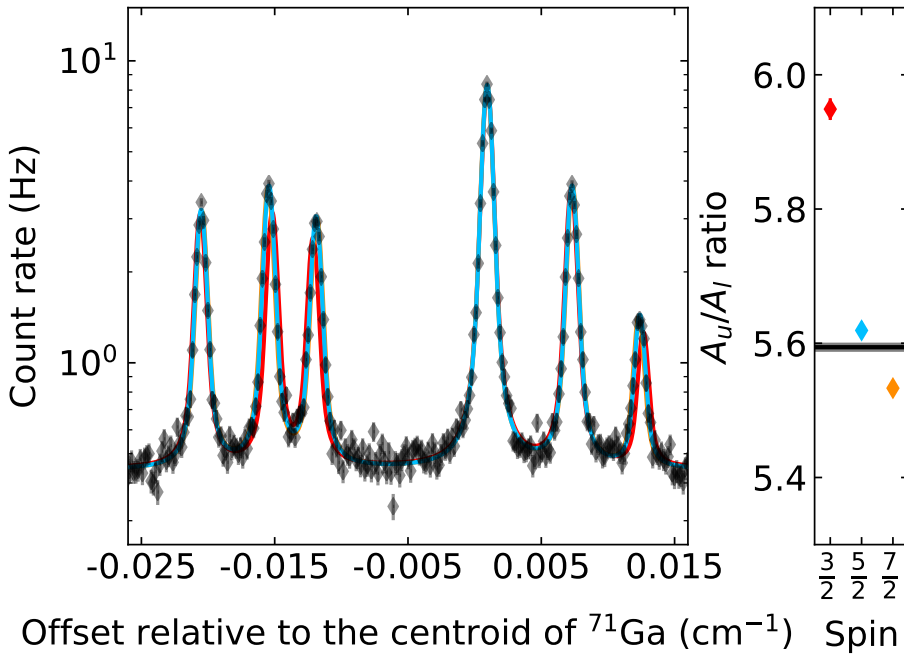


Figure 8.5: A simulated spectrum for the hyperfine structure of ^{83}Ga , fitted assuming $I = 3/2$ (red), $5/2$ (blue), and $7/2$ (orange) spin assignments. A corresponding A -hyperfine-parameter ratio analysis analogous to that displayed in Figure 6.2 for $^{75,79,81}\text{Ga}$, is displayed on the right-hand side. The estimated time required to experimentally replicate this simulation spanning 0.042 cm^{-1} with 258 steps is ~ 21 hours.

parameter predictions are needed for ^{84g}Ga with its $I = 0$ spin-state prediction. Isotope shifts for both isotopes assume a continuation of the kink observed for $^{81,82}\text{Ga}$ [Pro12] at the $N = 50$ shell closure, with a zero isomer shift between the two long-lived states in ^{84}Ga .

One major problem with the analysis of ^{81}Ga in Chapter 6.1 revolved around the inability to distinguish between the $I = 5/2$ and $7/2$ spin assignments from an A -hyperfine parameter analysis. The simulation presented in Figure 8.5 thus, is representative of the time required to discern between these two assignments with a $\geq 3\sigma$ difference with respect to their distances of their A -hyperfine parameter ratios from the accepted value for ^{71}Ga , assuming an $I = 5/2$ ground-state persists. Roughly 2.5 ISOLDE experimental shifts would be required, which within a standard CRIS experiment would present as an achievable measurement; the noticeable A -hyperfine parameter ratio difference for $I = 3/2$

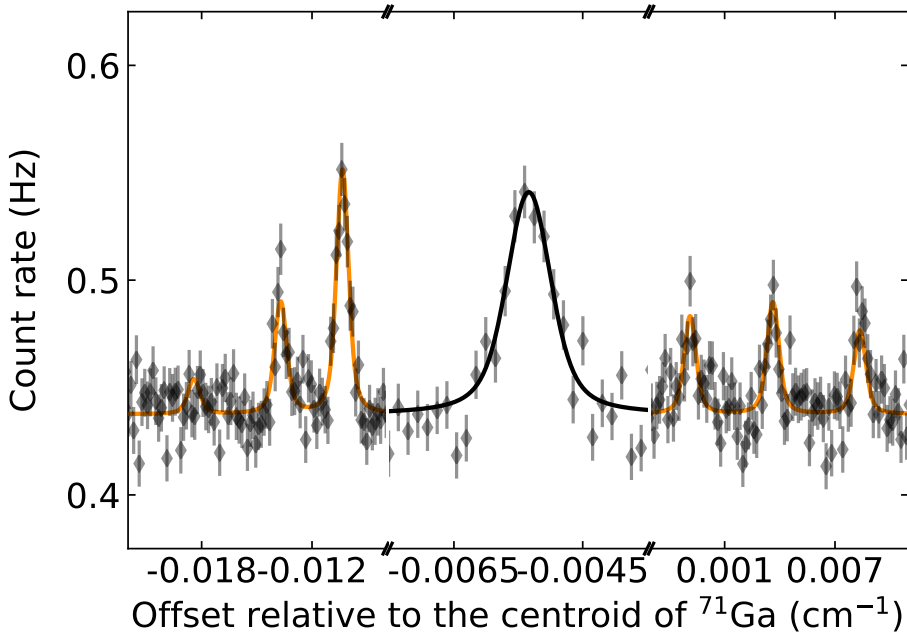


Figure 8.6: A simulated spectrum for the hyperfine structure of ^{84}Ga , fitted assuming an $I = 0$ (black) ground state and an $I = 3$ isomer state. The estimated time required to experimentally replicate these simulations spanning: 0.004 cm^{-1} for ^{84g}Ga with 27 steps is ~ 27 hours, and 0.028 cm^{-1} for ^{84m}Ga with 186 steps is ~ 186 hours.

means that there should be no problem in either ruling out or confirming this spin assignment in the aforementioned time. While the absence of available structural information for ^{83}Ga makes these statements rather tentative, it is likely some indication of its hyperfine structure would nonetheless manifest within the frequency region displayed in Figure 8.5. Regarding ^{84}Ga in Figure 8.6, the large reduction in its production yield from Table 8.2 has a profound effect on its hyperfine-spectroscopy measurement; although ~ 4 experimental shifts would be sufficient to measure the ground state, ≥ 20 experimental shifts would be required to confirm the existence of a purported long-lived isomer state. Like the case for ^{61}Ga in Chapter 8.1.1, this timescale for ^{84m}Ga would undoubtedly be unjustifiable for a CRIS experiment, especially considering the quoted background levels for an $A = 84$ isotope beam in Table 8.2 is likely to be considerably higher. This could however, provide further motivation for the experimental characterisation of the new neutron-converter setup at the ISOLDE facility, as already detailed in Chapter 3.1.1. Potentially improving

the StB ratio by up to a factor of 5 with respect to gallium-isotope beams [Ram19b], this would not only quicken the measurement times for obtaining the hyperfine spectra of $^{83,84}\text{Ga}$ in Figures 8.5 and 8.6, but also allow for successful measurements of $^{85,86}\text{Ga}$ and beyond [Coc13a].

8.1.3 Additional ionisation schemes for performing CRIS on gallium isotopes

The 417.2-nm transition detailed in Figure 3.5 for atomic gallium has been extensively studied in several laser-spectroscopy works [Che10b; Che10a; Man11a; Pro12; Che12a; Far17]. However as evidenced in Sections 6.1, 6.3.2, and 6.3.3, a convoluted spin-assignment analysis for the odd- A isotopes near $N = 50$, as well as the prevalence of compact hyperfine structures in $^{80,82}\text{Ga}$, have stymied their nuclear-structure interpretations and discussions. With every likelihood of these problems manifesting in the hyperfine structures for the heavier gallium species, it is intuitive to explore other feasible transition schemes which would allow for the easier extraction of their hyperfine observables, providing unambiguous clarity for any subsequent nuclear-structure interpretations. Thus, an extension of Figure 3.5 is given in Figure 8.7, highlighting other possible ionisation routes which are considered the most feasible⁵ to study neutral gallium at the CRIS experiment. The next couple of sections will discuss these transition schemes in more detail and elaborate on the laser-setup criteria needed to produce the required wavelengths. Unlike those presented previously in Chapters 8.1.1 and 8.1.2, simulations shown in this subchapter are not representative of the CRIS-experiment capabilities because the laser-ionisation, and subsequently the total experimental efficiency, remains unknown for these new transitions. Instead, the y -axis for these simulated hyperfine structures is represented by a normalised intensity with respect to the strongest observed transition.

The $ns\ ^2S_{1/2}$ series

Initially exciting to any of the other $^2S_{1/2}$ levels in neutral gallium provides an obvious starting point for the search of an alternative transition scheme. With respect to the $t_{1/2} = 5.7(6)$ ns lifetime for the $5s$ state [Lin81], those for the $6s$, $7s$, and $8s$ levels are considerably longer: 19(2) ns, 49(5) ns, and 80(10) ns respectively [Car86]. Longer excited-state lifetimes not only result

⁵Christine Steenkamp [Ste17] from Stellenbosch University in South Africa is thanked enormously for her valuable insights and discussions regarding this topic.

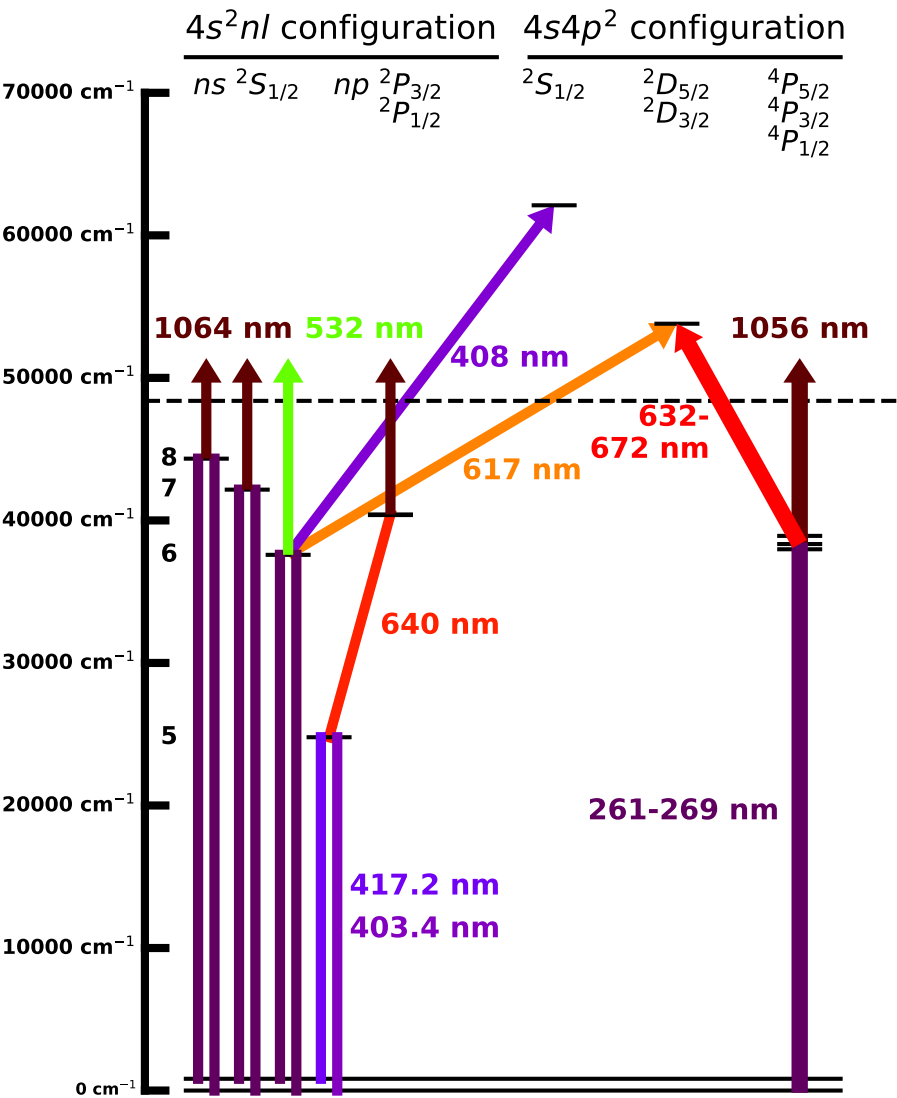


Figure 8.7: Additional transition schemes that could be used for measuring gallium isotopes at the CRIS experiment, starting from the $4p\ ^2P_{\frac{3}{2}, \frac{1}{2}}$ doublet. Numbers placed at the side of $ns\ ^2S_{1/2}$ levels represent the n number for that particular state; wavelengths required for transitions involving $n = 6, 7$, and 8 are stated later in the text.

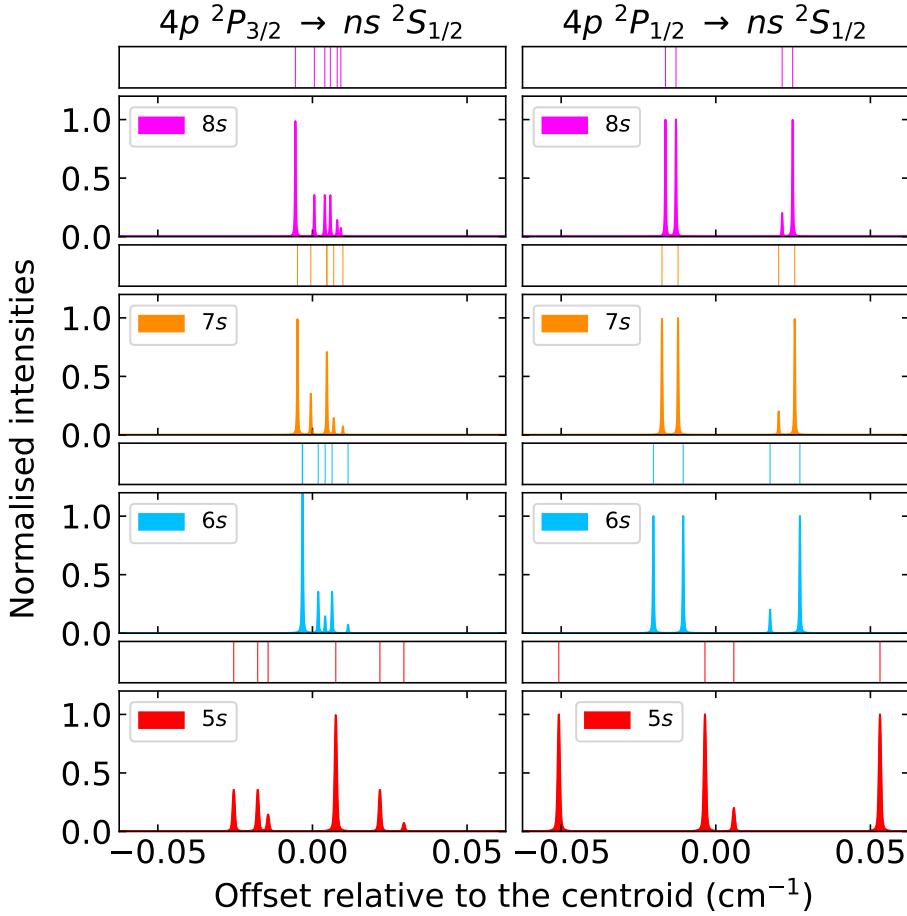


Figure 8.8: Simulated spectra for the hyperfine structure of ^{71}Ga , for all allowed transitions between $4p\ ^2P_{3/2}$ (left column), $4p\ ^2P_{1/2}$ (right column) $\rightarrow$ $5s, 6s, 7s, 8s\ ^2S_{1/2}$. The lines above each subplot allow for an easier display of the hyperfine-resonance positions. For the latter three upper levels, hyperfine parameters are taken from Ref. [Saf06]; the rest are taken from this thesis.

in narrower natural linewidths⁶, but also have the advantage of allowing for a greater flexibility concerning the manipulation of the laser-light pulses; for the gallium case, a better temporal delay could be applied for the 1064-nm light. This would help to eliminate the presence of lineshape distortions which

⁶Using (3.5) with the lifetimes given in the text: $\delta\nu_n(5s) \sim 28\text{ MHz}$, $\delta\nu_n(6s) \sim 9\text{ MHz}$, $\delta\nu_n(7s) \sim 3\text{ MHz}$, and $\delta\nu_n(8s) \sim 2\text{ MHz}$.

were previously observed in Chapter 4.4 for the 2015 IS571 experiment; this methodology had previously been done at the CRIS setup for copper and francium isotopes [Gro17d]. Figure 8.8 shows the resulting ^{71}Ga hyperfine spectrum that can be obtained for each of the possible transitions to the $ns\ ^2S_{\frac{1}{2}}$ series from the $4p\ ^2P_{\frac{3}{2},\frac{1}{2}}$ doublet. Although the 20 MHz Gaussian contribution from the beginning of Chapter 8.1 is still assumed, Lorentzian contributions are derived from the natural linewidths calculated in footnote #3 on p. 186, with an additional 10 MHz to account for any power-broadening effects.

Even though the ^{71}Ga hyperfine structures using the $J = 3/2 \rightarrow J' = 1/2$ schemes⁷ in Figure 8.8 are able to be fully resolved, they unfortunately become more collapsed with the involvement of higher n values, inevitably due to their progressively smaller A -hyperfine parameters [Saf06]. They would thus, be untenable for measuring collapsed gallium-isotope hyperfine structures, such as ^{80g}Ga and ^{82}Ga . It also explains why excitations to $n \geq 9$ states have not been considered, although the increasing difficulty of producing high-intensity ≤ 220 -nm UV light is also a limiting factor. While generally-wider ^{71}Ga hyperfine structures are observed in all of the featured $J = 1/2 \rightarrow J' = 1/2$ transition schemes⁸ in Figure 8.8, their impossibility for allowing electric-quadrupole-moment extractions and reduced sensitivity to the nuclear spin make them undesirable with respect to finding an improved gallium-transition scheme that would help to complete the aims set out in Ref. [Coc13a]. They could however, be used in conjunction with other transition schemes in order to help constrain the extraction of the hyperfine parameters; this is discussed further in the next subsection. If chosen for study, the involved wavelengths, as mentioned in the footnotes, can be obtained using a similar laser setup as described in Chapter 3.3.2 to produce 417.2-nm light, involving either frequency-tripling or frequency-quadrupling procedures where appropriate.

Completing the ionisation process if exciting to either the $7s$ or $8s$ levels can be achieved with just one additional 1064-nm step. For the $6s$ state however, this would not be sufficient for surpassing the IP; while 532-nm light could be used instead, this is not recommended because of the large cross-section for two-photon non-resonant ionisation of isobaric rubidium for what wavelength. Two autoionising states are highlighted in Figure 8.7 that could be used to further excite to: either the $4s4p^2\ ^2S_{1/2}$ level using 408-nm light, or the $4s4p^2\ ^2D_{\frac{5}{2},\frac{3}{2}}$ doublet using ~ 617 -nm light. Both represent forbidden transitions (for 408 nm: because of a two-electron jump, and for 617 nm: because of the $S \rightarrow D$ transition), thus requiring high-power laser-light densities. While no further

⁷For $J = 3/2 \rightarrow J' = 1/2$: 272.0-nm, 241.9-nm, and 229.8-nm light is required for $n = 6$, $n = 7$, and $n = 8$ respectively.

⁸For $J = 1/2 \rightarrow J' = 1/2$: 266.0-nm, 237.1-nm, and 225.5-nm light is required for $n = 6$, $n = 7$, and $n = 8$ respectively.

information is known about the $^2S_{1/2}$ state, the $^2D_{\frac{5}{2}, \frac{3}{2}}$ doublet is rather broad and is known to couple with the continuum [Con81; Buu88]. It is therefore recommended that any future offline studies into finding an alternate transition scheme for gallium at the CRIS setup should further investigate the usability of this autoionising state. 617-nm light can be produced fundamentally with a dye-laser using DCM dissolved in ethanol, analogous to the production of 639-nm light required for the previous three-step scheme discussed in Chapter 3.3.2.

The $^4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$ series

Another possibility to consider would be a transition to the $4s4p^2\ ^4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$ triplet, from the $4s^24p\ ^2P_{\frac{3}{2}, \frac{1}{2}}$ doublet. With respective lifetimes of: 2.6(1) μs , 4.2(4) μs , and 3.5(2) μs for the aforementioned triad of states [Buu88], their natural linewidths as calculated from (3.5) would effectively be negligible. Not only would this help in reducing the overall linewidth resolution, but it would also be sufficient to nullify the lineshape asymmetry as documented in Chapters 4.4.2 and 4.4.3. Although their forbidden⁹ natures would require high-power laser-light densities for sufficient saturation to occur, Ref. [Buu88] details a study in which these transition had been partially resolved using a dye-laser setup. Additionally, recent offline studies at the CRIS setup probed an analogue $5p\ ^2P_{3/2} \rightarrow 5s5p^2\ ^4P_{5/2}$ transition (at 283.7 nm) in neutral indium¹⁰ [Gar18]. Therefore based on both of these investigations, there should be no difficulty in being able to test these gallium transitions during an offline study at the CRIS setup. It is possible to produce the required 260-269-nm UV light using similar techniques utilised for the aforementioned 283.7-nm light in Ref. [Gar18]: with a similar laser setup for 417.2-nm light during the 2017 IS571 experiment as described in Chapter 3.3.2, along with nonlinear crystals for frequency-tripled light. With this setup, a total resolution of ~ 50 MHz can ultimately be obtained.

Expected ^{71}Ga hyperfine structures for all valid $4s^24p\ ^2P_{\frac{3}{2}, \frac{1}{2}} \rightarrow 4s4p^2\ ^4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$ transitions are presented in Figure 8.9; 20 MHz Gaussian and 10 MHz Lorentzian contributions are assumed, remembering that the natural linewidth can be taken as practically zero. Compared to those in Figure 8.8, wider structures are observed which span a frequency space of > 20 GHz. Thus, it might be necessary to perform scans initially scan in a broadband mode in order to establish the resonance positions; especially when considering any future

⁹This is because of an electron spin flip requirement; within neutral gallium, such transitions can be ~ 1000 less intensive than those that are allowed [Buu88].

¹⁰Both being part of the group 13 elements, the chemical similarity between gallium and indium mean that they should possess a similar ordering of atomic levels.

gallium experimental campaigns at the CRIS setup, its duration should take into account the total aggregate of frequency space that would inevitably be scanned. For reference, the study in Ref. [Gar18] took ~ 1 hour to scan a ~ 100 GHz range to measure the entire hyperfine structures of $^{113,115}\text{In}$ on a similar transition. Preferential consideration should also be given to the $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$ transition because of its increased sensitivity to the nuclear spin, which could help in making a firmer nuclear-spin assignment;

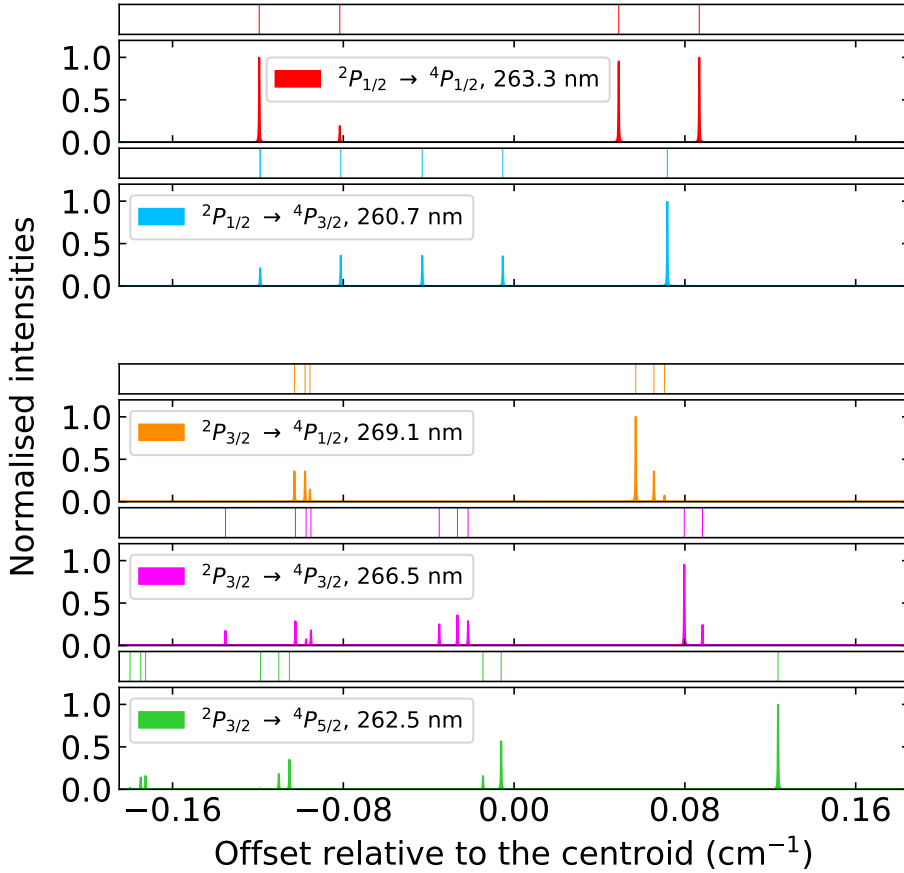


Figure 8.9: Simulated spectra for the hyperfine structure of ^{71}Ga , for all allowed transitions between $4p\ ^2P_{\frac{3}{2}, \frac{1}{2}} \rightarrow 4s4p^2\ ^4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$, provided in the legend for each subplot. The lines above each subplot allow for an easier display of the hyperfine-resonance positions. For the $^4P_{\frac{5}{2}, \frac{3}{2}, \frac{1}{2}}$ triplet state, hyperfine parameters are taken from Ref. [Buu88]; the rest are taken from this thesis.

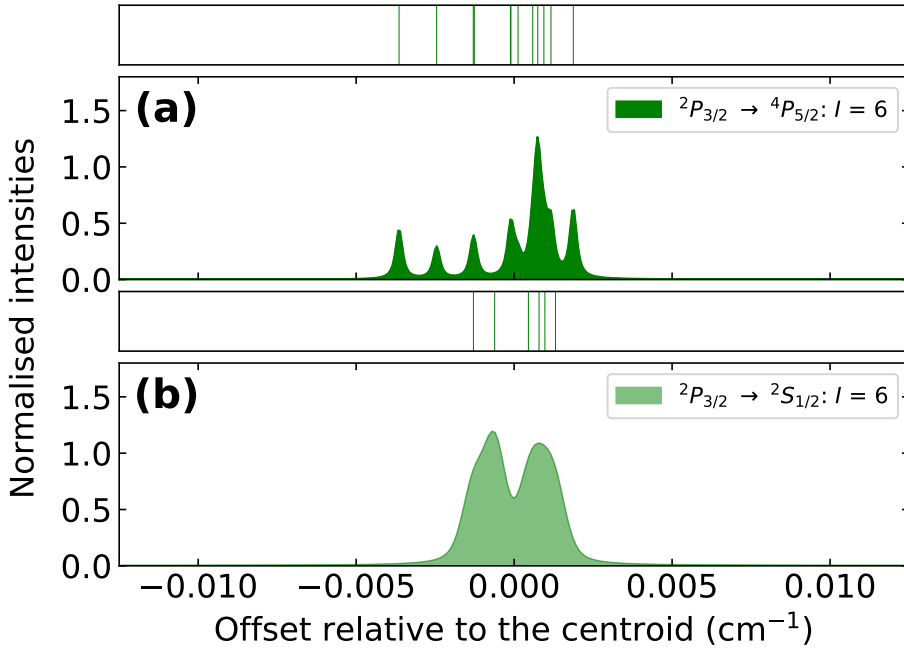


Figure 8.10: Simulated spectra for the hyperfine structure of ^{80}gGa , using the: (a) $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$, and (b) $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transitions. The lines above each subplot allow for an easier display of the hyperfine-resonance positions.

Figures 8.10 and 8.11 show how advantageous this could be for the cases of ^{80}gGa and ^{82}Ga respectively, compared to the previous 417.2-nm measurements made in this thesis. While still unable to be fully resolved, the observed ^{80}gGa structure would likely display more clearly-defined resonances, which would at least allow for further nuclear-spin assignments to be discounted. For ^{82}Ga , an unambiguous spin-value assignment could be made because of the unique numbers of expected transitions for each of the suspected $I = 1, 2$, and 3 spin assignments (see Chapter 2.3.3); if any long-lived isomerism is present, this could also be easily established.

If these transitions are considered for any future CRIS experimental campaigns, it is highly recommended to also measure a transition to one of the $ns\ ^2S_{1/2}$ series in conjunction, such that both transitions share the same initial state. This methodology has already been utilised in Ref. [Gar18]; not only would the constraining of the hyperfine parameters belonging to the shared initial state help to better predict the location of the larger ^4P structure, their hyperfine

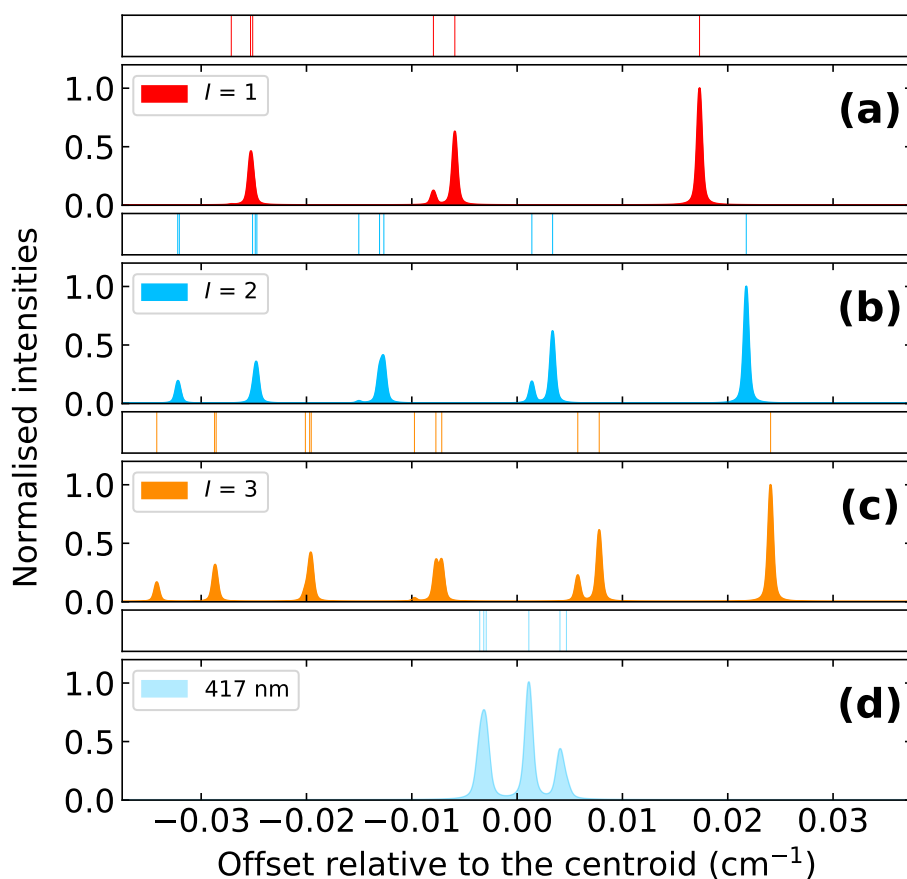


Figure 8.11: Simulated spectra for the hyperfine structure of ^{82}Ga , using the: (a,b,c) $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$ assuming the $I = 1, 2$, and 3 spin assignments, and (d) $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ transitions, assuming an $I = 2$ spin assignment. The lines above each subplot allow for an easier display of the hyperfine-resonance positions.

parameters would also be extracted with a better accuracy. Although the simulations of these transitions look rather promising for using to better study the nuclear structure of gallium isotopes, it is rather unfortunate in Figure 8.7 that subsequent direct ionisation requires 1056-nm light. At present, this cannot be produced with a sufficient intensity by detuning fundamental 1064-nm light from a Nd:YAG laser. Exciting to an autoionising state, such as the previously mentioned $^2D_{\frac{5}{2},\frac{3}{2}}$ doublet which would require 632-672-nm laser

light, is possible. This can be produced from a similar dye-laser setup used to produce 639-nm light in Chapter 3.1.2, with the use of DCM dye. Wavelengths beyond 660 nm however, would require the addition of DMSO in order to red-shift the dye [Wil19]; greater efficiency losses should also be expected considering the fluorescence intensity from DCM dye peaks at ~ 620 nm [Bir91; Bon04]. Overall, offline testing should be performed not only to determine the viabilities of these transition schemes with respect to successful gallium hyperfine-structure measurements at the CRIS setup, but also to search for other potential autoionising states.

8.1.4 Future decay spectroscopy of the two long-lived states in ^{80}Ga

As mentioned already in the introduction to Chapter 6, the decay spectroscopy of purified laser-ionised ^{80}Ga beams for the ground and isomer states could help to provide additional evidence of shape coexistence near ^{78}Ni , as already confirmed in Refs. [Got16; Yan16b]. It also aims to continue the β -decay,

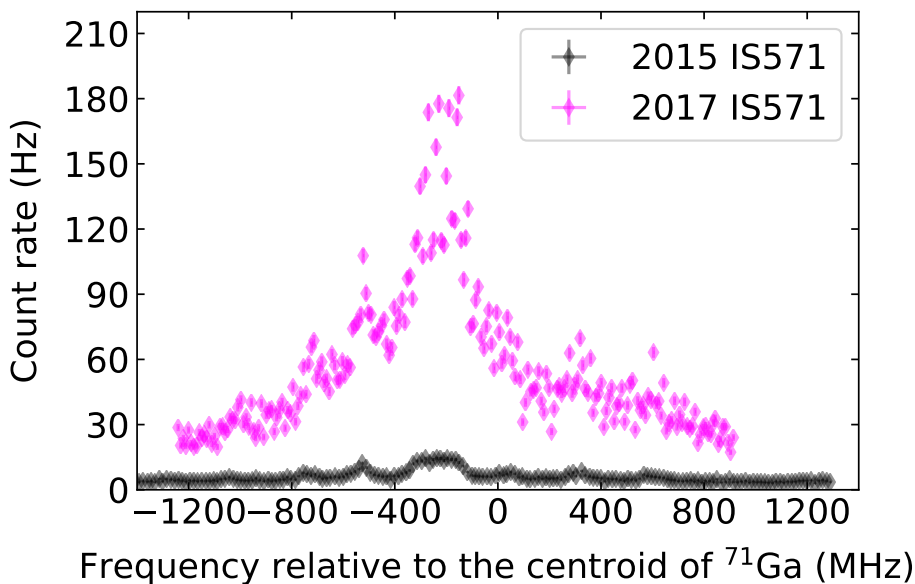


Figure 8.12: Representative hyperfine-spectrum measurements for the entire structure of ^{80}Ga taken during the: 2015 IS571 (black), and 2017 IS571 (magenta) experiments.

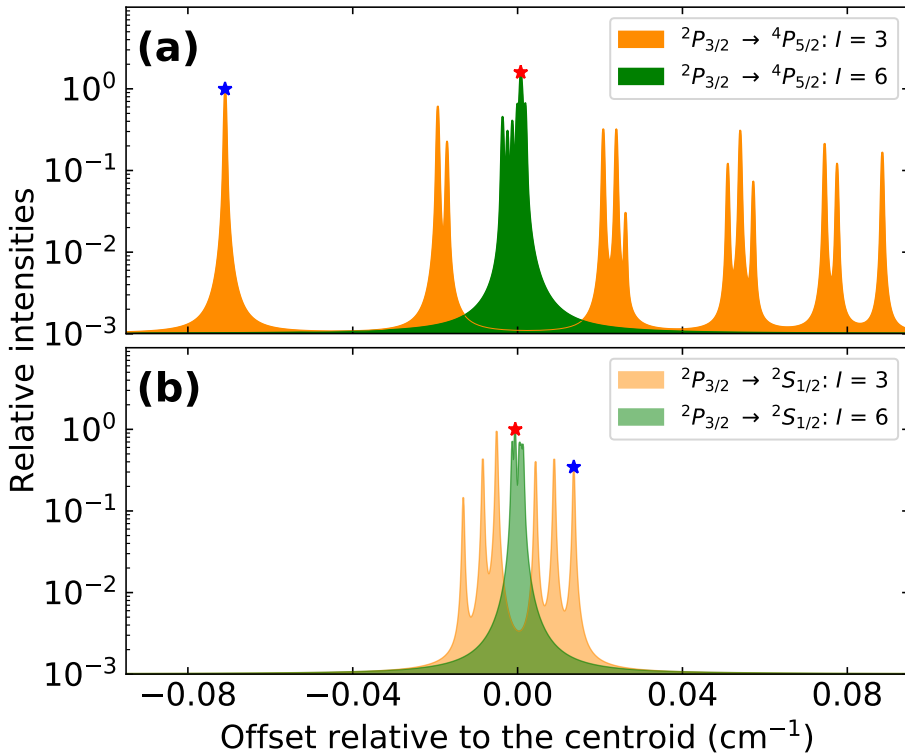


Figure 8.13: Overlaid hyperfine-spectrum simulations of ^{80g}Ga (green) and ^{80m}Ga (orange), using the: (a) $4p\ ^2P_{3/2} \rightarrow 4s4p^2\ ^4P_{5/2}$ (full colour), and (b) $4p\ ^2P_{3/2} \rightarrow 5s\ ^2S_{1/2}$ (shaded) transitions. The red and blue stars on each spectrum highlight the frequency locations in which the purest ground- and isomer-state beams can be obtained, respectively.

γ -decay, and conversion-electron spectroscopy studies already performed at the ALTO facility near IPN Orsay [Ver13; Got16]. One major hindrance in these aforementioned shape-coexistence studies had been the $\sim 10\%$ purity that could only be achieved for the $I = 3$ isomer-state beam. Disentangling the decay spectra for each state would not only help to complete the spectroscopic picture and help identify their exact feeding patterns, it would also allow for the determination of their half-lives independent from each other. New feeding patterns to the 0_2^+ intruder state in ^{80}Ge could also be additionally identified; at present, it is thought that only a 1764 keV γ transition (from a level at 2403 keV) feeds into this intruder state [Got16].

The CRIS setup is already capable of producing laser-ionised isotope beams for $^{80g,m}\text{Ga}$; this is evidenced in the obtained spectra for both IS571 experiments in Figure 8.12. With ground-state count rates of $\sim 15 \text{ s}^{-1}$ and $\sim 180 \text{ s}^{-1}$ reported for the 2015 and 2017 experiments respectively, this represents a factor of ~ 12 improvement at the CRIS setup during the two-year time period. For producing pure-state beams using the CRIS technique, this can simply be done by detuning the 417.2-nm laser light to be on-resonance with one of either the ground- or isomer-state peaks. It is also worth mentioning however, that the use of the $4p\ ^2P_{3/2} \rightarrow 4s4p\ ^2\ ^4P_{5/2}$ transition would offer a greater separation between the two hyperfine structures of $\geq 2 \text{ GHz}$ (compared to $\sim 500 \text{ MHz}$ for the previous transition), thus corresponding to a greater purity for any pure-state beams. Figure 8.13 graphically shows these differences; while a 99% purity could theoretically be achieved using the current transition scheme, this increases to at least $\sim 99.9\%$ if the proposed transition scheme is considered instead. This would be greatly beneficial for any future decay-spectroscopy studies performed at the CRIS setup involving $^{80g,m}\text{Ga}$ isotope beams. Choosing a suitable DSS however, is essential because although the DSS2.0 discussed in Chapter 3.3.4 is available, it would not be able to cope with the inevitable build-up of longer-lived ^{32}Ge and ^{33}As daughter nuclei from β decay and β -delayed neutron emission events (longest-lived: ^{79}As with $t_{1/2} = 9.01(15)$ minutes [Cun53]). A tape-transport system is more preferable because it would facilitate the removal of any long-lived contamination before its progressive accumulation would begin to hinder any decay-spectroscopy analysis. Such a setup had previously been used to attempt pure-state ^{80}Ga decay-spectroscopy studies during the 2017 IS571 experiment with the TATRA setup [Mat16; Ven17a; Ven17b], however repeated vacuum-related failures prevented any useful data collections. It is hoped that further development work on a suitable tape-transport system can be done, to facilitate future ^{80}Ga decay spectroscopy at the CRIS setup.

8.2 For francium isotopes

Compared to elemental gallium, a better experimental efficiency can be assumed for francium due to not only the general ease in which alkali(ne earth) elements with low ionisation potentials can be neutralised, but also a reduced population spread between the low-lying ground states because of their recombination patterns [Ver19b]. A total efficiency of $\sim 1/100$ is considered, which is sensible considering this has already been achieved for hyperfine-spectroscopy measurements of ^{202}Fr [Fla13b]. Voigt profiles are likewise assumed, however with narrower linewidths compared to those seen in gallium because of its reduced natural linewidth from the excited $8p\ ^2P_{3/2}$ state with $t_{1/2} = 83.5(15)$ ns [Aub04]. Based on the most recent francium-isotope experiment [Wil17b;

Wil17a], 20 MHz Gaussian and 15 MHz Lorentzian contributions are assumed. Additionally, associated contamination levels only consider isobaric radium because any other relevant species are either produced with too low yields at the ISOLDE facility, or too short-lived to prominently feature in francium-isotope measurements. The ISOLDE beamgate could also be utilised to further suppress longer-lived background levels; the use of a 10 ms beamgate every 2.4 s could theoretically reduce isobaric radium by a factor of 240 in any francium-isotope beam, and this is also considered. In accordance with the α -decay spectroscopy analysis in Chapter 7.2.3, any simulated isomer states have been suppressed by a factor of 5.2 with respect to the ground state.

8.2.1 ^{214,218}Fr: Characterising the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration

While the g -factor measurement of ^{214g}Fr documented in **Article II** suggested an incredibly pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration for an $I = (1)$ spin assignment, further studies are needed to investigate whether this shell-model prediction is solely confined or not. It is already suspected that this configuration predominantly explains not only its similarly-lived isomer state with $t_{1/2} = 3.35(5)$ ms [Tor68], but also for ^{218m}Fr [Bud14].

The j angular momenta for the orbitals involved in the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration provides a rather special case which is further explained in Appendix A.7: for all possible nuclear-spin states, empirical g factors as calculated from (2.17) will be invariant with respect to each other. Furthermore, their consequently similar A -hyperfine parameters can provide a useful first estimate for locating the isomer state in ²¹⁴Fr. Figure 8.14 shows their probable locations with either an $I = 8$ or $I = 9$ spin assignment, assuming a similar ground-state structure as observed in scan #1 of Figure 7.5. In analogy with the negligible isomer shift observed for the ground and isomer states in ²¹⁰Bi [Pea00], a zero isomer shift has been assumed for the case of ²¹⁴Fr. Overall, Figure 8.14 depicts that any high-spin isomer that is partnered with an $I = 2$ ground state would most likely feature well within the frequency-space ranges of the measurements shown in Figure 7.5. However as already discussed in Chapter 7.1.2, there was no such indication of its existence within statistical uncertainties. Another argument that should be considered is the assumption of similar hyperfine structures exhibited for both ^{214m}Fr and ^{218m}Fr, which is reasonable given their similar g factors to the empirical prediction for a $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration as depicted in Figure 7.10. From Ref. [Bud14], a multiplet separation spanning > 20 GHz for ^{218m}Fr is consistent with the scenario given in Figure 8.14(a); the isomer-state separation predicted in Figure 8.14(b) is insufficient. Thus, the rest of the simulations presented in this section

Table 8.3: Expected $^{214g,218g}\text{Fr}$ yields based on target-team measurements at the ISOLDE facility using UC_x target material, alongside background-limit estimates for isobaric radium. See Ref. [Bal19] regarding the compilation details of this data.

A	Fr Yield (μC^{-1})	Ra Yield (μC^{-1})
214g	9.4×10^2	9.0×10^6
218g ¹¹	4.3×10^4	9.0×10^6

assumes an $I = 1$ ground-state hyperfine structure for ^{214}Fr . Consequently, it also means that the frequency ranges used for the measurements in Figure 7.5 during the 2014 IS471 experiment were most likely not wide enough for the high-spin isomer to be observed. Based on the predicted locations in Figure 8.14(a), extending the chosen frequency ranges by ≤ 500 MHz would have been sufficient to confirm its presence.

Figure 8.15 displays a simulated spectrum for the full $^{214g,m}\text{Fr}$ structure based

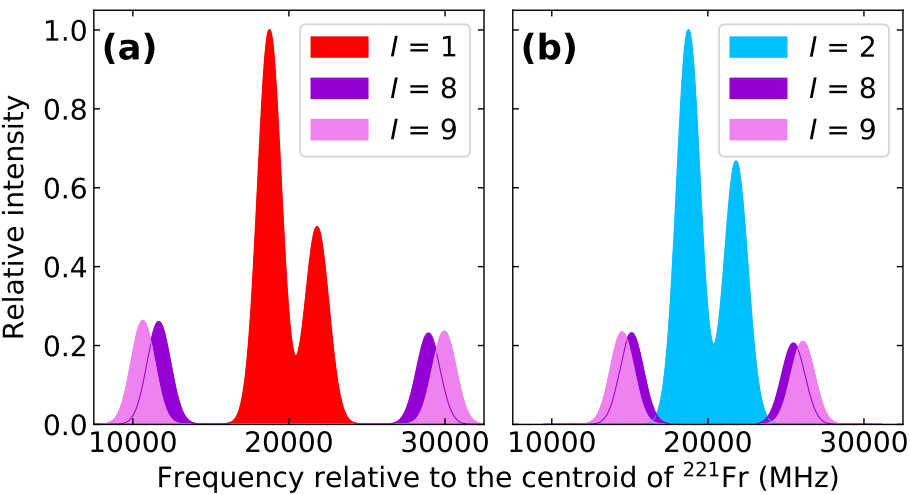


Figure 8.14: Simulated spectra for the hyperfine structure of ^{214m}Fr ($I = 8$ and 9 spin assignments; purple and pink), using the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition assuming: (a) $I = 1$ (red), and (b) $I = 2$ (blue) spin assignments for the ground state.

¹¹An upper estimate is made for the expected ^{218}Ra yield, based on those for ^{214}Ra and ^{220}Ra in which yield information using a UC_x target is available.

on the yield estimates in Table 8.3, as well as assuming ground- and isomer-state spin assignments of $I = 1$ and $I = 8$ respectively. A -hyperfine parameters for ^{214g}Fr are taken from Table 7.2, while B -hyperfine parameters are deduced from:

$$\frac{B}{Q_s} = 84.01\text{MHz}/b, \quad (8.1)$$

derived from many-body calculations using a CCSD_{t3} treatment [Sah15], with the theoretical electric-quadrupole moment calculation from Ref. [Yan18a]. The same methodology is used for modelling both A - and B -hyperfine parameters for ^{214m}Fr . Even by utilising the improved resolution now available at the CRIS setup in Figure 8.15, this would still be insufficient for obtaining a fully-resolved structure of ^{214g}Fr . Nonetheless, the observation of sub-structure present in the left multiplet would still allow for an electric-quadrupole moment measurement of

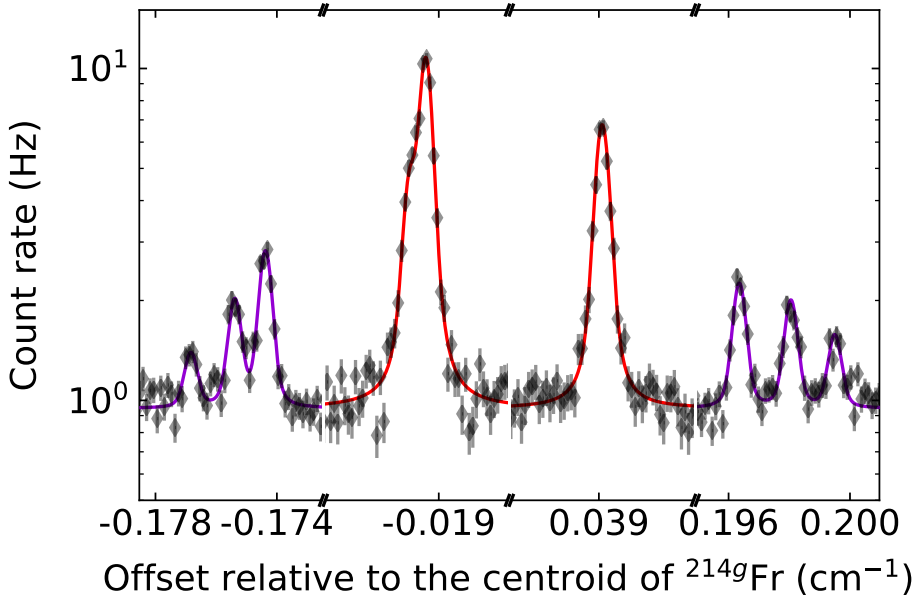


Figure 8.15: A simulated spectrum for the hyperfine structure of ^{214}Fr , fitted assuming an $I = 1$ (red) ground state and an $I = 8$ (purple) isomer state. The estimated time required to experimentally replicate these simulations spanning: 0.006 cm^{-1} for ^{214g}Fr with 103 steps is ~ 1.5 hours, and 0.006 cm^{-1} for ^{214m}Fr with 103 steps is ~ 3.5 hours.

this nuclear state; a near-zero value, corresponding to a spherical nuclear shape, would be expected in terms of the shell-model description due to the proximity to $N = 126$ and doubly-magic ^{208}Pb . Based on the theoretical $Q_s = +11.9 \text{ efm}^2$ value from Ref. [Yan18a], a $B(8p \ ^2P_{3/2}) \sim 10 \text{ MHz}$ hyperfine parameter represents the resolution limit that would be needed, albeit unobtainable at present, for a clear identification of each hyperfine resonance. With respect to the isomer state, a fully-resolvable structure is expected with the current available resolution, regardless of its nuclear-state spin assignment. The total time required to measure both states depicted in Figure 8.15 could be achieved well within the timeframe of one ISOLDE experimental shift.

Considering the similarities between the g factors for ^{214g}Fr and ^{218m}Fr as discussed beforehand, a re-measurement of the latter nuclear state would also be beneficial for further studying the influence of a $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ in other relevant even- A francium isotopes. Likewise using the tabulated yields provided in Table 8.3, Figure 8.16 shows the hyperfine structures of $^{218g,m}\text{Fr}$ that are

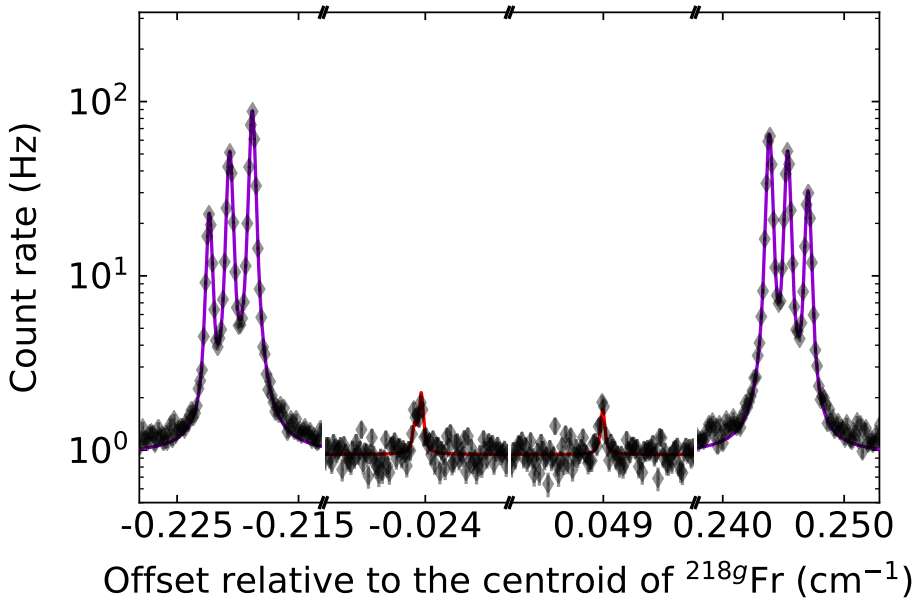


Figure 8.16: A simulated spectrum for the hyperfine structure of ^{218}Fr , fitted assuming an $I = 1$ (red) ground state and an $I = 8$ (purple) isomer state. The estimated time required to experimentally replicate these simulations spanning: 0.015 cm^{-1} for ^{218g}Fr with 209 steps is ~ 3.5 hours, and 0.015 cm^{-1} for ^{218m}Fr with 209 steps is ~ 7 hours.

expected to be exhibited. A -hyperfine parameters have been taken from Ref. [Bud14] and are assumed to be the same for both the $I = 1$ and $I = 8$ ground and isomer states, due of the special nature of the aforementioned pn coupling as discussed in Appendix A.7; similar B -hyperfine parameters to ^{214g}Fr are also assumed. The ground state is additionally suppressed by a factor of e^5 to account for half-life-related decay losses arising from a $t_{1/2} = 1.3(5)$ ms [Ewa82] lifetime, assuming the same Nd:YAG-laser system (with a 200 Hz duty cycle) that provided 1064-nm light for the 2014 IS471 experiment in Chapter 3.3.2, is used. It is remarked that this lifetime likely represents the limit of short-lived isotope production at ISOL facilities. While the hyperfine structures for the two long-lived states in ^{218}Fr could easily be measured within a single ISOLDE experimental shift, analogous to the case of ^{214}Fr beforehand, there would be significant challenges in being able to extract, and subsequently interpret, any information for the ground state. The use of a duty cycle > 200 Hz (possibly from another laser setup) would thus, undoubtedly help in mitigating any decay-related ^{218g}Fr losses at CRIS, required for successful measurements.

8.3 Summary

Although the contents of this thesis have provided further nuclear-structure insights with regards to some of the most intriguing features present within the nuclide chart, there is still further work to be done. This final discussion chapter has provided additional suggestions for further studying gallium and francium isotopes within the remit of this thesis: from further pushing the limits of exoticity for accessing both neutron-deficient and neutron-rich gallium species, to completing the characterisation of francium isotopes residing in the near vicinity to $N = 126$ and doubly-magic ^{208}Pb . Most of these have been proven to be achievable by using the current configuration of the CRIS experimental setup, within sensible time limits.

Chapter 9

Conclusions

This thesis has detailed laser spectroscopy measurements of the gallium isotopes $^{65,67,69,71,75,79-82}\text{Ga}$ and the francium isotope ^{214}Fr , using the collinear resonance ionisation spectroscopy technique at the ISOLDE facility in CERN. Fundamental ground-state properties such as: magnetic-dipole moments, electric-quadrupole moments (except ^{214}Fr), changes in the mean-squared charge radii, and tentative/firm nuclear spin assignments were extracted. Where applicable, their proximity to three doubly-magic nuclei: ^{56}Ni , ^{78}Ni , and ^{208}Pb , have allowed for fresh insights into characterising their single-particle strengths and testing the validity of nuclear shell model principles.

With leading $\pi 2p_{3/2}$ configurations, good agreements between the magnetic-dipole and electric-quadrupole moments of $^{65,67,69}\text{Ga}$ with LSSM GXPF1 calculations provide evidence for a rather soft ^{56}Ni core. JUN45 calculations also provide a good comparison, however further theoretical insights are needed to explain why such calculations assuming $g_{s,eff} = 1.0g_{s,free}$, *id est* unquenched, agree favourably with odd- A $_{31}\text{Ga}$ g factors in the $N = 28 - 40$ region, compared with their neighbouring $_{29}\text{Cu}$ and $_{33}\text{As}$ isotones. Mirrored electric-quadrupole moment trends between the odd- A $_{29}\text{Cu}$ and $_{31}\text{Ga}$ isotope chains provide an excellent example of the behavioural expectations for the nuclear shell model.

An electromagnetic moment analysis of neutron-rich gallium species from ^{71}Ga onwards, reveals an ever-changing shell structure towards $N = 50$. A gradual filling of the $\nu 1g_{9/2}$ orbital leads to the migration of protons from the $\pi 2p_{3/2}$ orbital to the $\pi 1f_{5/2}$ orbital which culminates in their inversion and subsequent $I^\pi = 3/2^- \rightarrow 5/2^-$ spin change at ^{81}Ga . A kink in the change in the mean-square charge radii for the succeeding isotope ^{82}Ga inherently suggests a magic $N = 50$ shell closure. Although extensive analyses of ground-state hyperfine

spectra for $^{80,82}\text{Ga}$ have been conducted, their compact natures prevent any definitive statements regarding their exact structures.

The measurement of ^{214g}Fr with a half-life of 5 ms represents a new limit of accessibility for studying short-lived nuclei at the CRIS setup. Despite the unavailability of high-resolution techniques, its g factor for a likely $I = (1)$ assignment can be explained with a remarkably pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration. Coupled with the kink in its change in the mean-square charge radii after $N = 126$, the magical character of ^{208}Pb is suggested to be rather tenacious five protons away from $Z = 82$. Additional electric-quadrupole moment, firm spin assignment, and improved isotope shift sensitivity measurements are not only suggested to help refine the complete nuclear structure picture of ^{214}Fr , but also to shed light on the Z -dependence with the magnitude of the charge radii kink for $N = 127$ isotones.

Although these studies have brought fresh insights into the characterisations of doubly-magic isotopes, there is further work that can be done. Albeit paraphrased, the quote from Section 1: *a golden pinnacle of scientific endeavour is the continuous and thorough scrutiny of any given proposal or statement*, should be reminded of once again. Not only will further measurements of francium and gallium isotopes help complete their respective stories, but those in neighbouring isotope chains are also crucial for fully discerning the magical reaches of these double shell-closure cores. Together with pushing the capabilities of ISOL facilities, the ongoing advancements at the CRIS setup will help make these measurements possible, vital for the concerted efforts to (hopefully) complete the mosaic of nuclear structure understanding.

Appendix A

Appendices

A.1 Essential quantum-mechanical relations

Further insights regarding the electromagnetic-moment descriptions stated in Chapter 2.1.2, require the application of quantum mechanics; key principles are now provided in this appendix. This also acts as a prelude to Appendix A.4, detailing the intricate derivation of neutron-orbital admixture contributions for the ground-state g factor of ^{214}Fr , continuing the physics discussion originating from Chapter 7.3.

The Wigner-Eckhart theorem

For a given tensor operator T_n^k of rank k and order n which acts on two angular-momentum states J and J' , each with respective projections M and M' , the Wigner-Eckhart theorem [Wig27; Eck30] can be used to decompose this expression as a product of individual angular-momentum and geometric contributions:

$$\langle J, M | T_n^k | J', M' \rangle = (-1)^{J-M} \begin{pmatrix} J & k & J' \\ -M & n & M' \end{pmatrix} \langle J || T^k || J' \rangle, \quad (\text{A.1})$$

with the expression enclosed within parentheses representing the Wigner-3 j coefficient¹.

¹Along with various relations, the mathematical definition of the Wigner-3 j coefficient can be found in Ref. [Bri94].

Reduction of a tensor operator

Reduced matrix elements can be rather complex and difficult to evaluate. However if its associated tensor in (A.1) can be described as a tensor product of operators $T^k(1, 2) = [T^{k_1}(1) \otimes T^{k_2}(2)]$, which each act on different independent subsystems, then the reduced matrix element can be further reduced [Hey90]:

$$\begin{aligned} & \langle (j_1 j_2), J || T^k(1, 2) || (j'_1 j'_2), J' \rangle \\ &= \sqrt{(2J+1)(2J'+1)(2k+1)} \left\{ \begin{array}{ccc} j_1 & j_2 & J \\ j'_1 & j'_2 & J' \\ k_1 & k_2 & k \end{array} \right\} \langle j_1 || T^{k_1} || j'_1 \rangle \langle j_2 || T^{k_2} || j'_2 \rangle, \end{aligned} \quad (\text{A.2})$$

to a product of smaller, simpler reduced matrix elements. In this instance, both subsystems represent a nuclear state with spins J and J' , each formed from the coupling of two individual shell-model orbitals with spins j_1, j_2 and j'_1, j'_2 respectively. The expression enclosed within curly brackets represents the Wigner-9j coefficient².

Provided these newly-disentangled reduced matrix elements each involve only a single shell-model orbital, they can be numerically evaluated with relative ease. Those with $j = j'$ can be equated to the appropriate single-particle multipole moment via the reverse-application of (A.1); $\langle j || M1 || j \rangle$ for example, is associated with the single-particle magnetic-dipole moment for the j orbital involved. Otherwise, they can be related to the reduced-transition probabilities [Bau73]:

$$B(TL; j' \rightarrow j) = \frac{1}{2j'+1} |\langle j || T^L || j' \rangle|^2. \quad (\text{A.3})$$

for applicable $j \rightarrow j'$ (TL)-character transitions. Whenever possible, single-particle multipole moments and reduced-transition probabilities should be taken from isotopes in the nearby vicinity of the nuclear system under study; values can be borne from either experimental or theoretical works.

²Likewise, various relations and the mathematical definition of the Wigner-9j coefficient can be found in Ref. [Bri94].

A.2 Derivation of the single-particle Schmidt moments

Starting with the formalism given on the first line in (2.14), the magnetic-dipole moment operator is re-written with respect to all relevant good quantum numbers:

$$\mu = \langle (l, s), j, m_j = j | \boldsymbol{\mu}_z | (l, s), j, m_j = j \rangle, \quad (\text{A.4})$$

namely j , m_j , l , and s . By projecting (A.4) with respect to j and acknowledging the definition of $\boldsymbol{\mu}$ stated in (2.12), this leads to:

$$\begin{aligned} \mu &= \langle (l, s), j, j | \boldsymbol{\mu} \cdot \mathbf{j} | (l, s), j, j \rangle \frac{\langle (l, s), j, j | j_z | (l, s), j, j \rangle}{\langle (l, s), j, j | \mathbf{j} \cdot \mathbf{j} | (l, s), j, j \rangle} \\ &= \frac{1}{j+1} \langle (l, s), j, j | \boldsymbol{\mu} \cdot \mathbf{j} | (l, s), j, j \rangle \\ &= \frac{1}{j+1} \langle (l, s), j, j | g_l(\mathbf{l} \cdot \mathbf{j}) + g_s(\mathbf{s} \cdot \mathbf{j}) | (l, s), j, j \rangle. \end{aligned} \quad (\text{A.5})$$

The two dot products featuring in the final line of (A.5) can be evaluated by squaring the following expressions $\mathbf{s} = \mathbf{j} - \mathbf{l}$ and $\mathbf{l} = \mathbf{j} - \mathbf{s}$, respectively:

$$\begin{aligned} \mu &= \frac{1}{j+1} \langle (l, s), j, j | \frac{g_l}{2}(\mathbf{j} \cdot \mathbf{j} + \mathbf{l} \cdot \mathbf{l} - \mathbf{s} \cdot \mathbf{s}) + \frac{g_s}{2}(\mathbf{j} \cdot \mathbf{j} - \mathbf{l} \cdot \mathbf{l} + \mathbf{s} \cdot \mathbf{s}) | (l, s), j, j \rangle \\ &= \frac{g_l[j(j+1) + l(l+1) - s(s+1)] + g_s[j(j+1) - l(l+1) + s(s+1)]}{2(j+1)}. \end{aligned} \quad (\text{A.6})$$

Finally, the last consideration to make is the $s = \pm 1/2$ substitution in (A.6), for the spin angular momentum of an individual nucleon. As already explained in Chapter 2.1.1, its addition to the shell model splits a particular l level into a doublet state, with total angular momenta of: $j = l + 1/2$, and $j = l - 1/2$. Naturally, two expressions arise from (A.6):

$$\begin{aligned}
\mu(j = l + 1/2) &= \frac{1}{2(j+1)} \left[g_l[(2j-1)(j+1)] + g_s(j+1) \right] \\
&= g_l l + \frac{1}{2} g_s, \\
\mu(j = l - 1/2) &= \frac{1}{j+1} \left[\frac{g_l}{2} [j(2j+3)] + \frac{g_s}{2} (-j) \right] \\
&= \frac{j}{j+1} \left[g_l(l+1) - \frac{1}{2} g_s \right],
\end{aligned} \tag{A.7}$$

resulting in the single-particle Schmidt moment expressions which are equivalent to those in (2.16).

A.3 Compilations of changes in the mean-square charge radii across the nuclear landscape

The rather colourful $\langle r^2 \rangle$ and $\delta \langle r^2 \rangle$ overviews around the ^{56}Ni , ^{78}Ni , and ^{208}Pb regions (where applicable) in Figures 5.9, 6.13, and 7.3 have been made possible thanks to extensive laser- and optical-spectroscopy studies over the years. Table A.1 gives an overview of the associated works used to compile all involved $\delta \langle r^2 \rangle$; for isotopes in which two or more such values are reported, a weighted mean originating from either the $\delta \nu$ (same-transition measurements) or $\delta \langle r^2 \rangle$ (different-transition measurements) data set is used for their final values. Conversions to $\langle r^2 \rangle$ values have been done using the following formula:

$$\langle r^2 \rangle^{A'} = \langle r^2 \rangle^A + \delta \langle r^2 \rangle^{A,A'}, \tag{A.8}$$

using theoretical $\langle r^2 \rangle^A$ reference-isotope predictions compiled in Ref. [Fri04]. Chosen reference points are also catalogued in Table A.1 on the right-most side; for elements whereby the $\delta \langle r^2 \rangle$ values for its associated isotopes are unknown from optical experimental techniques, available data from Ref. [Fri04] are used instead. $\langle r^2 \rangle$ references have not been used for the isotope chains featuring in Figure 7.3 (in the vicinity of doubly-magic ^{208}Pb) because of a general lack

Table A.1: References which contain $\delta\langle r^2 \rangle$ for the isotope chains featuring in Figures 5.9, 6.13, and 7.3. See text for further information regarding $\langle r^2 \rangle$ references.

Element	References	$\langle r^2 \rangle$ reference from Ref. [Fri04]
²⁴ Cr	[Fri04]	—
²⁵ Mn	[Hey16]	⁵⁵ Mn
²⁶ Fe	[Fri04; Min16]	⁵⁶ Fe
²⁷ Co	[Fri04]	—
²⁸ Ni	[Ste80]	⁶⁰ Ni
²⁹ Cu	[Bis16; Gro17a]	⁶⁵ Cu
³⁰ Zn	[Xie19]	⁶⁸ Zn
³¹ Ga	[Pro12; Far17]	⁷¹ Ga
³² Ge	[Fri04]	—
³³ As	[Fri04]	—
³⁴ Se	[Fri04]	—
³⁵ Br	[Fri04]	—
³⁶ Kr	[Kei95]	⁸⁶ Kr
³⁷ Rb	[Thi81; Man11b]	⁸⁷ Rb
³⁸ Sr	[Buc90]	⁸⁸ Sr
⁷⁶ Os	[Ang13; Hir17a]	—
⁷⁷ Ir	[Ver06]	—
⁷⁸ Pt	[Hil92; Le 99; Hir17b]	—
⁷⁹ Au	[Krö88; Wal89; Sav90; Pas94]	—
⁸⁰ Hg	[Ulm86; Mar18a; Sel18]	—
⁸¹ Tl	[Bar13; Bar17a]	—
⁸² Pb	[Ans86; Din87; Dut91; De 07]	—
⁸³ Bi	[Pea00; Bar16; Bar17b; Bar18]	—
⁸⁴ Po	[Kow91; Coc11; Sel13; Fin15]	—
⁸⁵ At	[Tei17; Cub18; Bar19]	—
⁸⁶ Rn	[Bor87]	—
⁸⁷ Fr	[Coc85; Duo87; Lyn14; Bud14]	—
	[Lyn16; Far16; Wil17b]	—
⁸⁸ Ra	[Wan12; Lyn18]	—
⁸⁹ Ac	[Tei17; Fer17; Ver19c]	—
⁹⁰ Th	[Käl89; Thi18; Saf18]	—

of these measurements in the trans-lead region [Coc17]. This explains why its landscape only displays the changes in the mean-square charge radii, in comparison to the other two (⁵⁶Ni and ⁷⁸Ni) regions mentioned in this section.

A.4 ν configuration-mixing calculations for the g factor of ^{214}gFr

While the g -factor analysis for ^{214}gFr presented in Chapter 7.3 and **Article II** revealed its nuclear structure can be described by a rather pure $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ configuration, discerning the exact nature of the additional contributions required remains an open question. One of the main focus points in **Article II** involved the role of the neutron orbitals, assuming a completely pure $(\pi 1h_{9/2}^5)_{9/2^-}$ proton configuration. Not only will this appendix describe the calculation details that lead to the conclusions stated in the aforementioned article, it will also provide additional calculations in order to provide a more general outlook. It is remarked that an uncertainty analysis for the upcoming calculations will not be given. This is due to a combination of: the lack of uncertainties for single-particle estimates in Chapter 7.3, the somewhat complicated nature of the following calculations, and the difficulty of ultimately representing them graphically.

This configuration-mixing exercise assumes the extended neutron model space displayed in Figure 7.9, encompassing the: $\nu 2g_{9/2}$, $\nu 1i_{11/2}$, and $\nu 2g_{7/2}$ shell-model orbitals. For this triad, the total ^{214}gFr wavefunction can be described by:

$$\begin{aligned} |^{214}\text{gFr}\rangle = & a |[(\pi 1h_{9/2}^5)(\nu 2g_{9/2})]\rangle + b |[(\pi 1h_{9/2}^5)(\nu 1i_{11/2})]\rangle \\ & + c |[(\pi 1h_{9/2}^5)(\nu 2g_{7/2})]\rangle, \end{aligned} \quad (\text{A.9})$$

with a , b , and c each denoting the amplitude for their respective configurations and are bound by the normalisation condition $|a|^2 + |b|^2 + |c|^2 = 1$. Even though Skyrme-Hartree-Fock (SHF) and RMF calculations predict a ~ 2 MeV energy gap between the $\nu 2g_{9/2}$ and $\nu 2g_{7/2}$ orbitals [God13], their spin-orbit-partner status warrants the inclusion of the latter in any configuration-mixing investigation because $M1$ -type transitions that connect between them can often non-linearly contribute to the total magnetic-dipole moment [Ari54]. Even small bestowals can have profound ramifications; the g factor of ^{51}Ca for example, has been found to alter by as much as 20% with just a 1% $M1$ -type contribution between the $\nu 1p_{3/2}$ and $\nu 1p_{1/2}$ orbitals [Gar15b; Gar15a]. Thus, any such occurrences should be included in any treatment. Utilising the traditional definition defined in (2.14), the state described in (A.9) has a magnetic-dipole moment of:

$$\begin{aligned}
 \langle ^{214}\text{Fr}|\mu|^{214}\text{Fr}\rangle &= a^2 \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]|M1|[(\pi 1h_{9/2})(\nu 2g_{9/2})]\rangle \\
 &\quad + b^2 \langle [(\pi 1h_{9/2})(\nu 1i_{11/2})]|M1|[(\pi 1h_{9/2})(\nu 1i_{11/2})]\rangle \\
 &\quad + c^2 \langle [(\pi 1h_{9/2})(\nu 2g_{7/2})]|M1|[(\pi 1h_{9/2})(\nu 2g_{7/2})]\rangle \\
 &\quad + 2ac \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]|M1|[(\pi 1h_{9/2})(\nu 2g_{7/2})]\rangle,
 \end{aligned} \tag{A.10}$$

which includes only the matrix elements that can be connected by an $M1$ transition because they have non-zero amplitudes. All displayed configurations in (A.10), which for the expression in (A.10) result in a negative parity and thus, any declaration of the total spin made in this appendix will assume this type of parity. Two assumptions have additionally been made for (A.10): that the aforementioned coefficients are numerically real, and that only the final valence $\pi 1h_{9/2}$ proton contributes to the total magnetic-dipole moment, by virtue of $g(\pi 1h_{9/2}^5) = g(\pi 1h_{9/2}^1)$ from (2.15). Evaluating the matrix elements associated with a^2 , b^2 , and c^2 , simply amount to calculating the g factor for the relevant pn configuration by using the additivity relation as described in (2.17). Alternatively, the formalism given in Appendix A.1 can also be used; a proof-of-equivalence for the a^2 component in (A.10) is now shown for completeness.

It is first reminded that an $M1$ operator possesses a rank $k = 1$. If an $I^\pi = 1^-$ ground state is initially assumed, then the a^2 -associated magnetic-dipole-moment matrix element corresponds to a maximal spin projection such that $J = M = 1$ and $J' = M' = 1$:

$$\begin{aligned}
 &a^2 \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = M = 1 | M1 | [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J' = M' = 1 \rangle \\
 &= a^2 (-1)^0 \begin{pmatrix} 1 & 1 & 1 \\ -1 & 0 & 1 \end{pmatrix} \times \\
 &\quad \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 || M1 || [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J' = 1 \rangle \\
 &= \frac{a^2}{\sqrt{6}} \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 || M1 || [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J' = 1 \rangle,
 \end{aligned} \tag{A.11}$$

in which a reduced matrix element results by utilising the Wigner-Eckhart theorem, as defined in (A.1) Its further reduction to a product of smaller

reduced matrix elements can be done with the application of (A.2), with respect to the individual $M1 = M1^\pi \otimes \mathbb{1} + \mathbb{1} \otimes M1^\nu$ proton and neutron contributions:

$$\begin{aligned}
 & \frac{a^2}{\sqrt{6}} \langle [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J = 1 || M1 || [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J' = 1 \rangle \\
 &= \frac{a^2}{\sqrt{6}} \left[\langle [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J = 1 || M1^\pi \otimes \mathbb{1} || [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J' = 1 \rangle \right. \\
 & \quad \left. + \langle [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J = 1 || \mathbb{1} \otimes M1^\nu || [(\pi 1 h_{9/2})(\nu 2 g_{9/2})]; J' = 1 \rangle \right] \\
 &= \sqrt{\frac{27}{6}} a^2 \left[\left\{ \begin{array}{ccc} 9/2 & 9/2 & 1 \\ 9/2 & 9/2 & 1 \\ 1 & 0 & 1 \end{array} \right\} \langle \pi 1 h_{9/2} || M1^\pi || \pi 1 h_{9/2} \rangle \langle \nu 2 g_{9/2} || \mathbb{1} || \nu 2 g_{9/2} \rangle \right. \\
 & \quad \left. + \left\{ \begin{array}{ccc} 9/2 & 9/2 & 1 \\ 9/2 & 9/2 & 1 \\ 0 & 1 & 1 \end{array} \right\} \langle \pi 1 h_{9/2} || \mathbb{1} || \pi 1 h_{9/2} \rangle \langle \nu 2 g_{9/2} || M1^\nu || \nu 2 g_{9/2} \rangle \right]. \tag{A.12}
 \end{aligned}$$

Choosing to use the aforementioned expression for $M1$ in (A.12) is deliberate. With respect to the pn -configuration states, it shows exactly how the $M1$ operator acts on the individual proton- and neutron-orbital contributions: $M1^\pi$ for the protons only, and $M1^\nu$ only for the neutrons. Their associated reduced matrix elements can be equated to the relevant single-particle magnetic-dipole moment via a reversed application of the Wigner-Eckhart theorem in (A.1), thus representing a relatively straightforward evaluation. For those involving the unit operator, the quantum-mechanical relation $\langle j_1 | \mathbb{1} | j_2 \rangle = \sqrt{j_1} \delta_{j_1, j_2}$ can be utilised, where δ_{j_1, j_2} is the Kronecker delta function. In other words, a non-zero evaluation only occurs if such reduced matrix elements connect between alike nucleons. From this, the calculation in (A.12) can be continued:

$$\begin{aligned}
 &= \sqrt{\frac{27}{6}} a^2 \left[\left\{ \begin{array}{ccc} 9/2 & 9/2 & 1 \\ 9/2 & 9/2 & 1 \\ 1 & 0 & 1 \end{array} \right\} \langle \pi 1 h_{9/2} || M1^\pi || \pi 1 h_{9/2} \rangle \langle \nu 2 g_{9/2} || \mathbb{1} || \nu 2 g_{9/2} \rangle \right. \\
 & \quad \left. + \left\{ \begin{array}{ccc} 9/2 & 9/2 & 1 \\ 9/2 & 9/2 & 1 \\ 0 & 1 & 1 \end{array} \right\} \langle \pi 1 h_{9/2} || \mathbb{1} || \pi 1 h_{9/2} \rangle \langle \nu 2 g_{9/2} || M1^\nu || \nu 2 g_{9/2} \rangle \right]
 \end{aligned}$$

$$\begin{aligned}
 &= \sqrt{\frac{27}{6}} a^2 \times \frac{1}{45\sqrt{22}} \left[\frac{\mu_{\pi 1h_{9/2}}}{(-1)^{9/2-9/2} \begin{pmatrix} 9/2 & 1 & 9/2 \\ -9/2 & 0 & 9/2 \end{pmatrix}} \sqrt{10} \delta_{\pi 2g_{9/2}, \pi 2g_{9/2}} \right. \\
 &\quad \left. + \sqrt{10} \delta_{\pi 1h_{9/2}, \pi 1h_{9/2}} \frac{\mu_{\nu 2g_{9/2}}}{(-1)^{9/2-9/2} \begin{pmatrix} 9/2 & 1 & 9/2 \\ -9/2 & 0 & 9/2 \end{pmatrix}} \right] \\
 &= \sqrt{\frac{27}{132}} \frac{a^2}{45} \frac{\sqrt{110}}{3} \sqrt{10} [\mu_{\pi 1h_{9/2}} + \mu_{\nu 2g_{9/2}}] \\
 &= \frac{a^2}{9} [\mu_{\pi 1h_{9/2}} + \mu_{\nu 2g_{9/2}}] \\
 &= 0.314 \text{a}^2 \mu_N,
 \end{aligned} \tag{A.13}$$

whereby the penultimate line in (A.13) uses the single-particle magnetic-dipole moment values $\mu(\pi h_{9/2}) = +4.02 \mu_N$ [Coc85] and $\mu(\nu g_{9/2}) = -1.197 \mu_N$ [Sel14]. If the additivity relation in (2.17) is used instead, this yields:

$$\begin{aligned}
 \mu &= gI = \frac{1}{2} \left[\frac{4.02}{9/2} - \frac{1.197}{9/2} + \left(\frac{4.02}{9/2} + \frac{1.197}{9/2} \right) \frac{(9/2 \times 11/2) - (9/2 \times 11/2)}{(1 \times 2)} \right] \\
 &= \frac{2\mu_N}{18} [4.02 - 1.197] \\
 &= 0.314 \text{a}^2 \mu_N,
 \end{aligned} \tag{A.14}$$

giving an equivalent answer to that presented in (A.14). Therefore, both of the methods presented in (A.13) and (A.14) can be used to numerically evaluate the matrix elements associated with a squared coefficient term, which for the expression in (A.10) are a^2 , b^2 , and c^2 . Matrix-element calculations associated with the latter two coefficients utilise the $\mu_{\nu 1i_{11/2}} = +0.695 \mu_N$ and $\mu_{\nu 2g_{7/2}} = +0.945 \mu_N$ single-particle values from ^{209}Pb in Ref. [Bau73]. Numerically evaluating the ac term is more involved, but can be done with the combined methodology in (A.11), (A.12), and (A.13):

$$\begin{aligned}
& 2ac \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = M = 1 | M1 | [(\pi 1h_{9/2})(\nu 2g_{7/2})]; J' = M' = 1 \rangle \\
&= 2ac(-1)^0 \begin{pmatrix} 1 & 1 & 1 \\ -1 & 0 & 1 \end{pmatrix} \times \\
&\quad \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 | | M1 | | [(\pi 1h_{9/2})(\nu 2g_{7/2})]; J' = 1 \rangle \\
&= \frac{2ac}{\sqrt{6}} \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 | | M1 | | [(\pi 1h_{9/2})(\nu 2g_{7/2})]; J' = 1 \rangle \\
&= \frac{2ac}{\sqrt{6}} \left[\langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 | | M1^\pi \otimes \mathbb{1} | | [(\pi 1h_{9/2})(\nu 2g_{7/2})]; J' = 1 \rangle \right. \\
&\quad \left. + \langle [(\pi 1h_{9/2})(\nu 2g_{9/2})]; J = 1 | | \mathbb{1} \otimes M1^\nu | | [(\pi 1h_{9/2})(\nu 2g_{7/2})]; J' = 1 \rangle \right] \\
&= \sqrt{\frac{27}{6}} 2ac \left[\begin{pmatrix} 9/2 & 9/2 & 1 \\ 9/2 & 7/2 & 1 \\ 1 & 0 & 1 \end{pmatrix} \langle \pi 1h_{9/2} | | M1^\pi | | \pi 1h_{9/2} \rangle \langle \nu 2g_{9/2} | | \mathbb{1} | | \nu 2g_{7/2} \rangle \right. \\
&\quad \left. + \begin{pmatrix} 9/2 & 9/2 & 1 \\ 9/2 & 7/2 & 1 \\ 0 & 1 & 1 \end{pmatrix} \langle \pi 1h_{9/2} | | \mathbb{1} | | \pi 1h_{9/2} \rangle \langle \nu 2g_{9/2} | | M1^\nu | | \nu 2g_{7/2} \rangle \right]. \tag{A.15}
\end{aligned}$$

A key difference between the last line of (A.15) with that of (A.12) is the $\langle \nu 2g_{9/2} | | \mathbb{1} | | \nu 2g_{7/2} \rangle$ matrix element attached with the $M1^\pi$ contribution. Due to the conditions of the Kronecker delta, this part is equivalent to zero and thus, only the (latter) $M1^\pi$ neutron term contributes to the admixture term. The other $\langle \nu 2g_{9/2} | | M1^\nu | | \nu 2g_{7/2} \rangle$ term can be numerically evaluated from (A.3), using a theoretical $B(M1; \nu 2g_{7/2} \rightarrow \nu 2g_{9/2}) = 0.482 \mu_N^2$ value for ^{209}Pb [Bau73]. Therefore for (A.15):

$$\begin{aligned}
& \sqrt{\frac{27}{6}} 2ac \left[\begin{pmatrix} 9/2 & 9/2 & 1 \\ 9/2 & 7/2 & 1 \\ 1 & 0 & 1 \end{pmatrix} \langle \pi 1h_{9/2} | | M1^\pi | | \pi 1h_{9/2} \rangle \langle \nu 2g_{9/2} | | \mathbb{1} | | \nu 2g_{7/2} \rangle \right. \\
&\quad \left. + \begin{pmatrix} 9/2 & 9/2 & 1 \\ 9/2 & 7/2 & 1 \\ 0 & 1 & 1 \end{pmatrix} \langle \pi 1h_{9/2} | | \mathbb{1} | | \pi 1h_{9/2} \rangle \langle \nu 2g_{9/2} | | M1^\nu | | \nu 2g_{7/2} \rangle \right]
\end{aligned}$$

$$\begin{aligned}
 &= \frac{2\sqrt{27}ac}{\sqrt{6}} \left[\left\{ \begin{array}{ccc} 9/2 & 9/2 & 1 \\ 9/2 & 7/2 & 1 \\ 0 & 1 & 1 \end{array} \right\} \langle \pi 1h_{9/2} || 1 || \pi 1h_{9/2} \rangle \langle \nu 2g_{9/2} || M1^\nu || \nu 2g_{7/2} \rangle \right] \\
 &= 3\sqrt{2}ac \left[\frac{1}{90} \sqrt{\frac{11}{2}} \times \sqrt{10} \times \pm \sqrt{8 * 0.482\mu_N^2} \right] \\
 &= \pm \frac{2ac}{3} \sqrt{\frac{11 \times 0.482\mu_N^2}{5}} \\
 &= \pm 0.687ac\mu_N,
 \end{aligned} \tag{A.16}$$

finally resulting in a numerical evaluation for the ac admixture term stated in (A.10), assuming an $I^\pi = (1^-)$ assignment. Calculating the same for an $I^\pi = (2^-)$ assignment can be done by simply following the same methodology in (A.15) and (A.16), but using the $J = M = 2$ and $J' = M' = 2$ projections. Overall for (A.10), the magnetic-dipole moment (and subsequent g -factor) descriptions for $I^\pi = (1^-)$ and (2^-) equate to:

$$\begin{aligned}
 &\frac{\langle ^{214}\text{Fr}; I^\pi = 1^- | \mu | ^{214}\text{Fr}; I^\pi = 1^- \rangle}{I} \\
 &= g(^{214}\text{Fr}; I^\pi = 1^-) = (0.314a^2 - 1.599b^2 + 1.984c^2 \pm 0.687ac)\mu_N,
 \end{aligned} \tag{A.17}$$

$$\begin{aligned}
 &\frac{\langle ^{214}\text{Fr}; I^\pi = 2^- | \mu | ^{214}\text{Fr}; I^\pi = 2^- \rangle}{I} \\
 &= g(^{214}\text{Fr}; I^\pi = 2^-) = (0.314a^2 - 0.193b^2 + 1.049c^2 \pm 0.387ac)\mu_N.
 \end{aligned} \tag{A.18}$$

With the g -factor relations for $^{214}\text{g}\text{Fr}$ now defined in (A.17) and (A.18) with respect to the nearby neutron orbitals, two scenarios will now be imagined: the first one looking at two-neutron-state mixing cases which had been assumed and discussed in **Article II**, and a second one looking at the generalised picture for all three neutron orbits. Where appropriate, contributions required for a specific g -factor scenario are expressed as a ratio of percentages, like: %/%(/%). While of course these assume a pure $(\pi 1h_{9/2}^5)_{I^\pi=9/2^-}$ contribution, they nonetheless quantify the role of the neutron orbitals in this g -factor analysis.

Scenario I: Two-neutron-state orbital mixing

In order to simplify the discussion, **Article II** assessed the ^{214}Fr g -factor composition under two separate scenarios by assuming in both (A.17) and (A.18): one, a zero contribution from the $\nu 1i_{11/2}$ orbital ($b = 0$) and two, a zero contribution from the $\nu 2g_{7/2}$ orbital ($c = 0$). Figure A.1 shows their graphical interpretations for the $I^\pi = (1^-)$ and (2^-) assignments, in which the application of the stringent normalisation condition allows these g -factor expressions to be plotted with respect to the occupation of a single neutron orbital in Figure A.1. For both cases, the $\nu 2g_{9/2}$ orbital represents the most logical choice, considering its immediate proximity after the $N = 126$ shell.

When the $\nu 2g_{7/2}$ orbital contributions are neglected, linear g -factor relations result (orange lines in Figure A.1) with respect to the $\nu 2g_{9/2}$ and $\nu 1i_{11/2}$ orbital contributions. Compared with the experimental g factors for $I^\pi = (1^-)$ and (2^-) (red and blue lines respectively), the neutron-orbital mixtures required

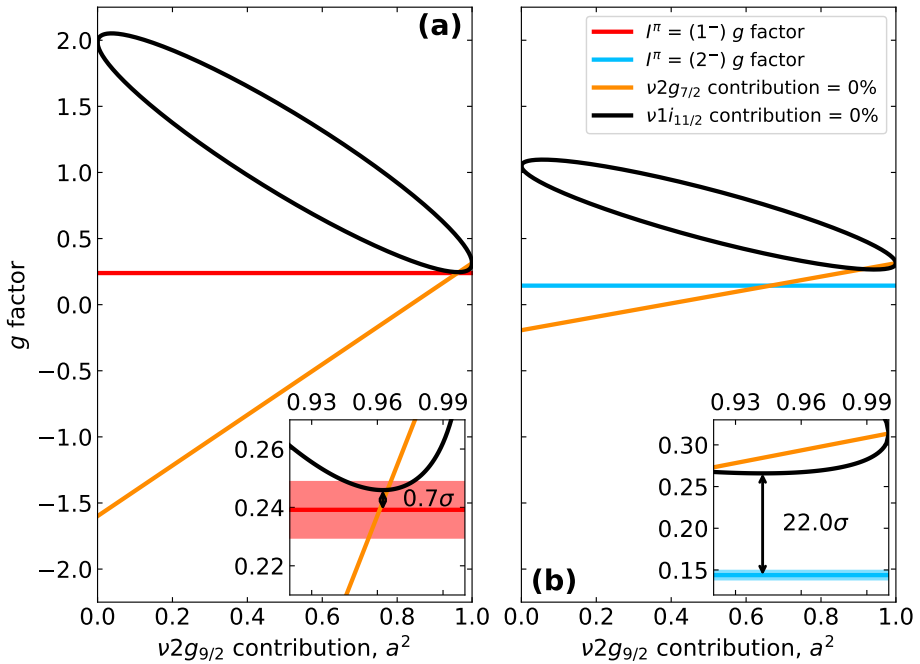


Figure A.1: Empirically calculated ^{214}Fr g -factor relations assuming: (a) $I^\pi = (1^-)$, and (b) $I^\pi = (2^-)$, for two-neutron-state mixing cases as defined in the text. The insets accentuate the local minimum for each g -factor scenario, with respect to the respective experimental values.

($\nu 2g_{9/2}/\nu 1i_{11/2}$) are analogous to those depicted in Figure 7.10: 96%/4%, and 66%/34%, required for the aforementioned spin assignments respectively. However if the $\nu 1i_{11/2}$ orbital contributions are assumed to be zero instead, then the g -factor relations appear elliptical (black curves in Figure A.1) because of the inclusion of a non-zero admixture term. The experimental g -factor values thus, may not necessarily be described by just one unique solution, such as the case for ^{51}Ca mentioned beforehand in Ref. [Gar15b]. Although the insets of Figures A.1(a) and A.1(b) reveal that the g factors obtained by the CRIS setup do not exactly correspond to valid solutions, the 0.7σ difference for the $I^\pi = (1^-)$ case is sufficiently close such that a 4% $\nu 2g_{7/2}$ admixture contribution could also explain the rest of its corresponding g factor. A 22.0σ gap for the $I^\pi = (2^-)$ case however, definitively excludes the possibility of admixture contributions contributing towards its total g factor. Nonetheless, this g -factor analysis for both spin assignment suggests their respective wavefunctions are dominated with the lone valence neutron residing in the $\nu 2g_{9/2}$ orbital, in agreement with LSSM calculations. These are the conclusions that have also been presented in **Article II** regarding the overall $^{214}\text{G}\text{Fr}$ g -factor discussion.

Scenario II: Three-neutron-state orbital mixing

Following from the previous two-neutron-state scenario, additional work has since been carried out in order to explore the likeliest contributions to the $^{214}\text{G}\text{Fr}$ g factor from all three neutron orbitals, without any simplifications. The ellipses presented in Figure A.2 for both $I^\pi = (1^-)$ and (2^-) spin assignments, now represent planes of general solutions for the experimental g factors presented in Table 7.5; a two-dimensional representation has been maintained by quantifying the $\nu 2g_{7/2}$ orbital contributions with a colour map. The validity³ of the two-neutron-orbital solution obtained in the previous section, with respect to these solution planes, add credence to the validity of this methodology. In general, configurations with a prevalent presence of $\nu 2g_{9/2}$ contributions only require a minimal influence from the $\nu 1i_{11/2}$ and $\nu 2g_{7/2}$ orbitals. The most predominant for $I^\pi = (1^-)$ and (2^-) require 98%/1%/1% and 70%/29%/1% ratios of $\nu 2g_{9/2}/\nu 1i_{11/2}/\nu 2g_{7/2}$ neutron contributions respectively; some degree of $\nu 1i_{11/2}$ orbital contributions are needed to explain the experimental g factor for both cases, for all possible solutions. Configurations involving minimal $\nu 2g_{9/2}$ orbital contributions are also mathematically plausible, however the consequent wavefunctions are unlikely to describe the ground state of ^{214}Fr , considering the closeness of the $I^\pi = (1^-)$ and (2^-) g factors near the empirical ($\pi 1h_{9/2} \otimes$

³The small deviation for the bottom-most $I^\pi = (1^-)$ solution in the inset of Figure A.2(a) is due to the assumption of zero $\nu 1i_{11/2}$ orbitals contributions that had been made in the previous section. This of course, shows up quite clearly under the three-neutron-mixing regime.

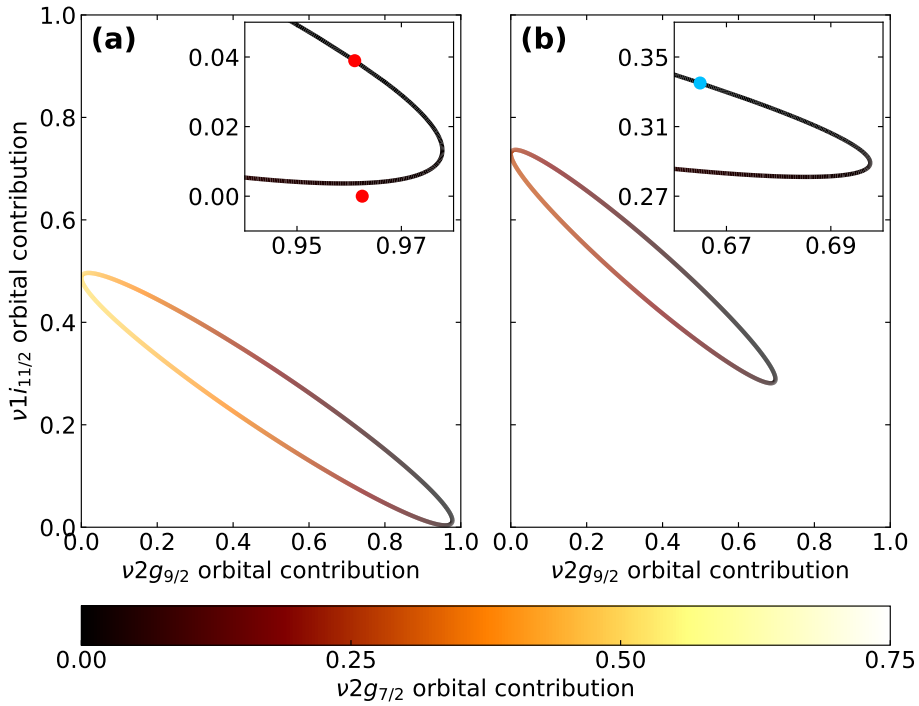


Figure A.2: General solution planes for experimental ^{214}gFr g -factor values assuming: (a) $I^\pi = (1^-)$, and (b) $I^\pi = (2^-)$, with respect to $\nu 2g_{9/2}$, $\nu l_{1/2}$, and $\nu 2g_{7/2}$ orbital contributions. The insets magnify the turning points associated with the purest $\nu 2g_{9/2}$ configurations; proposed solutions for each spin assignment mentioned in **Scenario I** are indicated by the coloured dots.

$\nu 2g_{9/2}$) g -factor value in Figure 7.10. Nonetheless, this statement cannot be ruled out at present and it further highlights the importance of measuring the electric-quadrupole moment of ^{214}gFr , as well as additional theoretical insights, in order to fully characterise its nuclear structure.

A.5 A spin-assignment-analysis method for hyperfine structures measured with low resolution

Analyses of low-resolution laser-spectroscopy measurements can amount to a rather arduous effort, because the inability to discern the exact quantity of observed peaks in a hyperfine spectrum can complicate spin-assignment analyses.

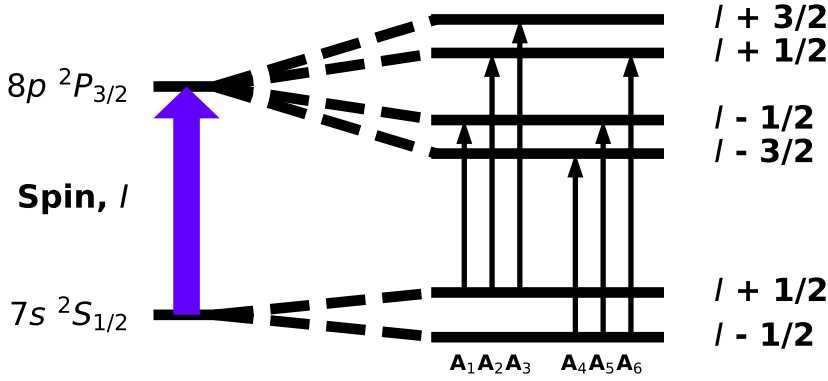


Figure A.3: A diagram of an example hyperfine spectrum for the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition in francium, applicable for an isotope with nuclear spin of $I \geq 3/2$. Each F -hyperfine state is labelled with respect to I .

However, it is possible to make tentative assignments by comparing the observed intensity ratio for each multiplet with theoretically-calculated values that are expected for certain spin assignments. An example will now be done for the case of francium isotopes studied under the $7s\ ^2S_{1/2} \rightarrow 8p\ ^2P_{3/2}$ transition in **Article II** and Chapter 7.

For familiarity, Figure A.3 shows the expected hyperfine transitions under the aforementioned scenario. Each are labelled (from left-most) A_{1-6} : A_1 , A_2 , and A_3 belong to the left multiplet, while A_4 , A_5 , and A_6 belong to the right multiplet. Their relative intensities can be calculated from the definition provided in (2.45), involving the use of Wigner- $6j$ coefficients which themselves can be related to the Racah W -coefficients:

$$\left\{ \begin{matrix} j_1 & j_2 & j_3 \\ j_4 & j_5 & j_6 \end{matrix} \right\} = (-1)^{j_1+j_2+j_4+j_5} W(j_1 j_2 j_5 j_4; j_3 j_6), \quad (\text{A.19})$$

with their algebraic definitions defined in Refs. [Bie52; Edm57; Bri94]. Although their full calculations look at first glance, cumbersome and complex, special cases exist which can dramatically reduce the algebra involved. The formulas that are relevant for this exercise can be found in Table II of Ref. [Bie52]. Based on these, along with (2.45), the calculations for A_{1-6} are now given individually:

$$\begin{aligned}
A_1 &= (2I+2)(2I) \left\{ \begin{array}{ccc} I+1/2 & I-1/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I+2)(2I) \left[(-1)^{2I+2} W(I+1/2, I-1/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I+2)(2I) \frac{(2)(1)(2I)(2I-1)}{4(2I)(I+1/2)(2I+2)(2)(3/2)(4)} \\
&= \frac{I(2I-1)}{12(I+1/2)},
\end{aligned} \tag{A.20}$$

$$\begin{aligned}
A_2 &= (2I+2)(2I+2) \left\{ \begin{array}{ccc} I+1/2 & I+1/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I+2)(2I+2) \left[(-1)^{2I+3} W(I+1/2, I+1/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I+2)(2I+2) \frac{(2I+3)(2I)(1)(2)}{4(I+1/2)(2I+2)(I+3/2)(3/2)(4)(2)} \\
&= \frac{I(I+1)}{3(I+1/2)},
\end{aligned} \tag{A.21}$$

$$\begin{aligned}
A_3 &= (2I+2)(2I+4) \left\{ \begin{array}{ccc} I+1/2 & I+3/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I+2)(2I+4) \left[(-1)^{2I+4} W(I+1/2, I+3/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I+2)(2I+4) \frac{(2I+4)(2I+3)(3)(2)}{4(2I+4)(I+3/2)(2I+2)(4)(3/2)(2)} \\
&= \frac{I+2}{2}.
\end{aligned} \tag{A.22}$$

$$\begin{aligned}
A_4 &= (2I)(2I-2) \left\{ \begin{array}{ccc} I-1/2 & I-3/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I)(2I-2) \left[(-1)^{2I} W(I-1/2, I-3/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I)(2I-2) \frac{(3)(2)(2I-1)(2I-2)}{4(2I-2)(I-3/2)(2I)(2)(3/2)(4)} \\
&= \frac{I-1}{2},
\end{aligned} \tag{A.23}$$

$$\begin{aligned}
A_5 &= (2I)(2I) \left\{ \begin{array}{ccc} I-1/2 & I-1/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I)(2I) \left[(-1)^{2I+1} W(I-1/2, I-1/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I)(2I) \frac{(2I+2)(2I-1)(2)(1)}{4(I-1/2)(2I)(I+1/2)(3/2)(4)(2)} \\
&= \frac{I(I+1)}{3(I+1/2)},
\end{aligned} \tag{A.24}$$

$$\begin{aligned}
A_6 &= (2I)(2I+2) \left\{ \begin{array}{ccc} I-1/2 & I+1/2 & 1 \\ 3/2 & 1/2 & I \end{array} \right\}^2 \\
&= (2I)(2I+2) \left[(-1)^{2I+2} W(I-1/2, I+1/2, 1/2, 3/2; 1, I) \right]^2 \\
&= (2I)(2I+2) \frac{(2I+3)(2I+2)(2)(1)}{4((2I+2)(I+1/2)(2I)(4)(3/2)(2))} \\
&= \frac{(2I+3)(2I+2)}{12(2I+1)},
\end{aligned} \tag{A.25}$$

and are all expressed solely as a function of I . Denoting the left-hand and right-hand multiplets in Figure A.3 as S_1 and S_2 respectively, their relative intensities can be determined by summing up the appropriate individual relative intensities from (A.20), (A.21), (A.22), (A.23), (A.24), and (A.25):

$$\begin{aligned}
S_1 &= A_1 + A_2 + A_3 \\
&= \frac{I(2I-1)}{12(I+1/2)} + \frac{I(I+1)}{3(I+1/2)} + \frac{I+2}{2} \\
&= \frac{I(2I-1) + 8I(2I+2) + 3(2I+1)(I+2)}{6(2I+1)} \\
&= \frac{12I^2 + 18I + 6}{6(2I+1)} \\
&= \frac{6(2I+1)(I+1)}{6(2I+1)} \\
&= I+1,
\end{aligned} \tag{A.26}$$

$$\begin{aligned}
S_2 &= A_4 + A_5 + A_6 \\
&= \frac{I-1}{2} + \frac{I(I+1)}{3(I+1/2)} + \frac{(2I+3)(2I+2)}{12(2I+1)} \\
&= \frac{6(2I+1)(I-1) + 8I(I+1) + (2I+3)(2I+2)}{12(2I+1)} \\
&= \frac{24I^2 + 12I}{12(2I+1)} \\
&= \frac{12(2I+1)}{12(2I+1)} \\
&= I,
\end{aligned} \tag{A.27}$$

finally allowing the ratio between the left-hand multiplet (A.26), with respect to the right-hand multiplet (A.27), to be defined as:

$$S = \frac{S_1}{S_2} = \frac{I+1}{I}. \tag{A.28}$$

Remarkably, this represents a simple expression for calculating the expected intensity ratio between observed multiplet resonances. It also shows the

increasing similarity of hyperfine structures associated with higher values of I . Although this derivation is based on Figure A.3, (A.28) is valid for all I ; repeating the derivation for $I = 1/2$ or 1 for example, yields the same result.

A.6 A review of francium isotope-shift measurements used for a King-plot analysis

The $\delta\langle r^2 \rangle^{214,221}$ values presented in **Article II** had been calculated using the F_{422} and M_{422} factors determined from a King-plot analysis presented in Ref. [Lyn14]; a replication can be seen in Figure A.4(a). However since its publication, new/additional francium isotope-shift data are available for: $^{204g,m1,m2}\text{Fr}$, ^{205}Fr , $^{206g,m1,m2}\text{Fr}$, and ^{207}Fr (422.7 nm: Refs. [Lyn14; Lyn16; Wil17b], 718.0 nm⁴: Ref. [Vos15]). Thus, an updated version incorporating these additions is presented in Figure A.4(b); the two side-by-side subfigures allow for an easy comparison. Corresponding F_{422} and M_{422} atomic parameters

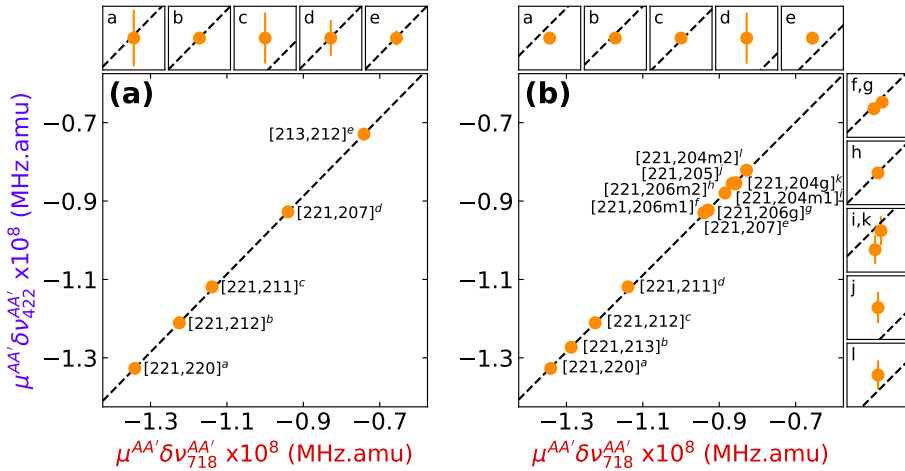


Figure A.4: A comparison of the King plots used for the extraction of F and M atomic factors associated with the 422.7-nm, based on: (a) original data from Ref. [Lyn14], and (b) to date, all available data. The insets belonging to a King plot provide a zoomed-in view for each data point with respect to the line of best fit. See text for the references of the new available data.

⁴Considering the isotope shifts reported in Ref. [Vos15] use ^{208}Fr as the reference, the value for $\delta \nu^{208,221}$ from Ref. [Coc85] has been added to each isotope shift so that they can be comparable those extracted from the 422.7-nm transition.

Table A.2: Extracted F_{422} and M_{422} factors resulting from the progressive addition of updated data and new analysis techniques.

	F_{422} (GHz fm $^{-1}$)	M_{422} (GHz amu)
Ref. [Lyn14]	−20.67(21)	+750(330)
Figure A.4(a); previous data	−20.667(214)	+750(334)
Figure A.4(b); updated data	−20.272(227)	−1626(474)
Updated data + corr.	−20.272(204)	−1626(141)

for the King plots shown in Figure A.4 are presented in Table A.2, calculated from the extracted gradients and intercepts using (2.44) and the methodology in Chapter 2.3.2. For the 718.0-nm transition, atomic-theory calculations [Dzu05] predict: $F_{718} = -20.766(208)$ GHz fm $^{-1}$, and $M_{718} = -85(113)$ GHz amu, which have been used for the $\delta\langle r^2 \rangle$ -value determinations in Ref. [Vos15].

One often-neglected consideration in King-plot analyses is discerning the degree of correlation between the gradient and intercept. Other KU Leuven Ph.D

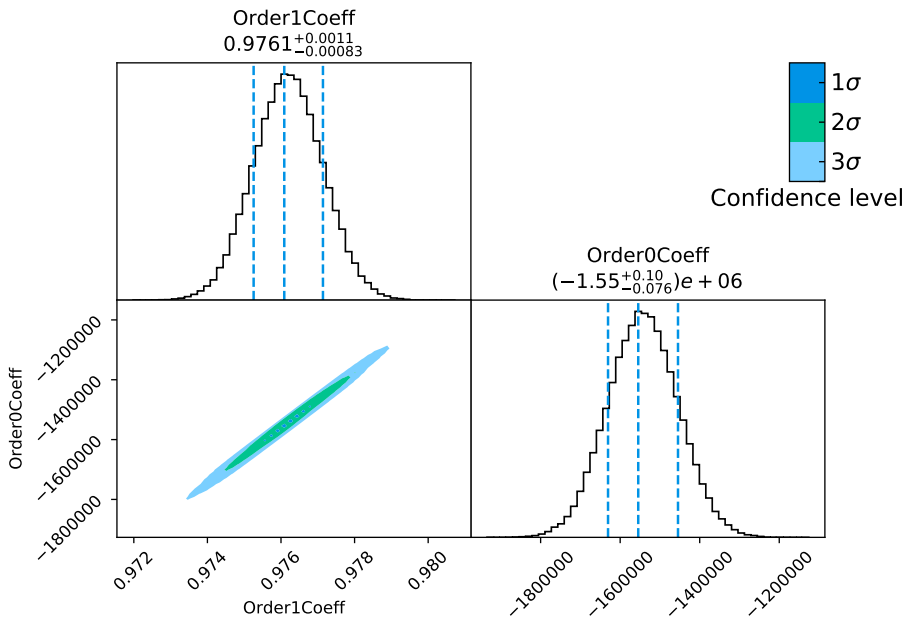


Figure A.5: A correlation plot between the gradient (named Order1Coeff) and intercept (named Order0Coeff) of the King plot presented in Figure A.4(b).

theses have already studied this for the copper [Gro17a] and argon [Gin19] isotope shifts. In line with their methodologies, a Bayesian statistical analysis has also been performed, involving a Bootstrapping method with a Monte-Carlo Markov Chain (MCMC) algorithm; further information regarding this technique can be found in Chapter 5.5.1 of Ref. [Gro17a]. Figure A.5 shows its results, in which the highly-correlated behaviour between the gradient and intercept in the bottom-left plot confirms the need to properly treat these manifestations. Subsequently-obtained F_{422} and M_{422} values are presented in the fourth line of Table A.2; while the aforementioned analysis does not modify their values, their total uncertainties have been reduced by $\sim 10\%$ and $\sim 70\%$ respectively.

To conclude, re-calculated $\delta\langle r^2 \rangle^{A,221}$ values are catalogued in Table A.3 for every extracted francium isotope shift at the CRIS setup, using the newly-updated F_{422} and M_{422} atomic parameters presented in the fourth line of Table A.2.

Table A.3: $\delta\langle r^2 \rangle$ comparisons between: previously-published and newly re-calculated CRIS values (second and third column), with TRIUMF values (fourth column).

A	$\delta\langle r^2 \rangle^{A,221}$		
	Old CRIS values	New CRIS values	TRIUMF values
202g	−1.5965(49)[176]	−1.5779(48)[161]	
202m	−1.5912(64)[175]	−1.5725(63)[161]	
203	−1.5301(3)[168]	−1.5131(3)[155]	
204g	−1.5710(49)[171]	−1.5576(48)[159]	−1.5560(2)[140]
204m1	−1.5773(49)[171]	−1.5640(48)[159]	−1.5579(2)[140]
204m2	−1.5130(49)[165]	−1.4984(48)[153]	−1.5050(2)[139]
205	−1.4744(49)[160]	−1.4619(48)[149]	−1.4737(2)[139]
206g	−1.4849(1)[160]	−1.4754(1)[150]	−1.4752(2)[139]
206m1	−1.4868(1)[160]	−1.4773(1)[150]	−1.4776(2)[139]
206m2	−1.4152(1)[153]	−1.4043(1)[143]	−1.4054(2)[139]
207	−1.3897(3)[149]	−1.3810(3)[141]	
211	−1.1708(49)[124]	−1.1686(49)[119]	
214g ($I = 1$)	−0.9500(45)[99]	−0.9513(45)[96]	
214g ($I = 2$)	−0.9623(46)[101]	−0.9638(46)[98]	
218m	−0.4009(49)[42]	−0.4015(49)[41]	
219	−0.2664(2)[28]	−0.2667(2)[27]	
220	−0.1338(49)[14]	−0.1340(49)[14]	
221	0	0	
229	+0.8940(49)[94]	+0.8930(49)[91]	
231	+1.0782(49)[114]	+1.0764(49)[109]	

Values quoted for ^{214g}Fr in Table 7.6 are taken from these new calculations. Previously-published CRIS-setup [Bud14; Lyn14; Lyn16; Far16; Wil17b] and TRIUMF⁵ [Vos15] values are also included for comparison. It is remarked that the inconsistency between the CRIS-setup and TRIUMF $\delta\langle r^2 \rangle^{A,221}$ values for $^{206g,m1,m2}\text{Fr}$, as reported in Ref. [Lyn16], is now rectified with the new CRIS-setup values. In future, the use of the new aforementioned F_{422} and M_{422} atomic parameters is strongly recommended for any francium isotope-shift measurements made on the 422.7-nm transition.

A.7 A special case for a pn coupling with $j_p = j_n$

While different empirical g factors calculated from (2.17) for a particular pn coupling are expected for each allowed state, a special case arises for when $j_p = j_n$. For the $(\pi 1h_{9/2} \otimes \nu 2g_{9/2})$ pn configuration associated with ^{214g}Fr in Chapter 7.3, its g factor according to (2.17) with $j_p = j_n = 9/2$ is:

$$\begin{aligned} g &= \frac{1}{2} \left[g_p + g_n + (g_p - g_n) \frac{9/2(9/2 + 1) - 9/2(9/2 + 1)}{I(I + 1)} \right] \\ &= \frac{1}{2} [g_p + g_n], \end{aligned} \quad (\text{A.29})$$

resulting in an expression which is completely independent of the total nuclear-state spin I , assuming this pn configuration can 100% describe the nuclear structure for that state. As an example for ^{214}Fr , this could mean that $g(^{214g}\text{Fr}_{I=(1)}) = g(^{214m}\text{Fr}_{I=(8)})$ for the ground and isomer states. Determining their expected A -hyperfine parameters can be done by considering the aforementioned statement, along with substituting (2.40) into (2.13) and labelling the ground and isomer states g and m respectively:

$$\begin{aligned} g_m I_m &= g_g I_g \frac{I_m A_m}{I_g A_g} \\ g_m A_g &= g_g A_m \\ A_g &= A_m. \end{aligned} \quad (\text{A.30})$$

Thus ultimately, the A -hyperfine parameters are expected to be similar for the $I = (1)$ and (8) states in ^{214}Fr ; they are in general, the same for all allowed I .

⁵These use the same isotope-shift values that feature in Figure A.4(b).

Bibliography

- [AlK96] J. S. Al-Khalili and J. A. Tostevin. “Matter Radii of Light Halo Nuclei.” *Physical Review Letters* **76** (1996) 3903–3906.
- [All14] J. M. Allmond *et al.* “High-precision $B(E2)$ measurements of semi-magic $^{58,60,62,64}\text{Ni}$ by Coulomb excitation.” *Physical Review C* **90** (2014) 034309.
- [Als16] M. F. Alshudifat *et al.* “Reexamining Gamow-Teller decays near ^{78}Ni .” *Physical Review C* **93** (2016) 044325.
- [Alt17] N. A. Althubiti *et al.* “Spectroscopy of the long-lived excited state in the neutron-deficient nuclides $^{195,197,199}\text{Po}$ by precision mass measurements.” *Physical Review C* **96** (2017) 044325.
- [And00] A. N. Andreyev *et al.* “A triplet of differently shaped spin-zero states in the atomic nucleus ^{186}Pb .” *Nature* **405** (2000) 430–433.
- [And02] A. N. Andreyev, K. Van de Vel *et al.* “Nuclear spins, magnetic moments and α -decay spectroscopy of long-lived isomeric states in ^{185}Pb .” *The European Physical Journal A - Hadrons and Nuclei* **14** (2002) 63–75.
- [And05] L-L. Anderson *et al.* “Identification of excited states in $^{31}_{61}\text{Ga}_{30}$: Mirror nuclei in the upper fp shell.” *Physical Review C* **71** (2005) 011303(R).
- [And17] A. N. Andreyev, A. E. Barzakh *et al.* “Shape-coexistence and shape-evolution studies for bismuth isotopes by in-source laser spectroscopy.” *CERN-INTC P-443-ADD-1* (2017).
- [Ang13] I. Angeli and K. P. Marinova. “Table of experimental nuclear ground state charge radii: An update.” *Atomic Data and Nuclear Data Tables* **99** (2013) 69–95.
- [Ans86] M. Anselment *et al.* “The odd-even staggering of the nuclear charge radii of Pb isotopes.” *Nuclear Physics A* **451** (1986) 471–480.

- [Ari54] A. Arima and H. Horie. “Configuration Mixing and Magnetic Moments of Odd Nuclei.” *Progress of Theoretical Physics* **12** (1954) 623–641.
- [Ata15] D. Atanasov *et al.* “Precision Mass Measurements of $^{129-131}\text{Cd}$ and Their Impact on Stellar Nucleosynthesis via the Rapid Neutron Capture Process.” *Physical Review Letters* **115** (2015) 232501.
- [Aub04] S. Aubin *et al.* “Lifetimes of the $9s$ and $8p$ levels of atomic francium.” *Physical Review A* **70** (2004) 042504.
- [Aud17] G. Audi *et al.* “The NUBASE2016 evaluation of nuclear properties.” *Chinese Physics C* **41** (2017) 030001.
- [Aut55] S. H. Autler and C. H. Townes. “Stark Effect in Rapidly Varying Fields.” *Physical Review* **100** (1955) 703–722.
- [Aye18] S. Ayet San Andrés. “Developments for Multiple-Reflection Time-of-Flight Mass Spectrometers and their Application to High-Resolution Accurate Mass Measurements of Short-Lived Exotic Nuclei.” *Ph.D. thesis, Justus-Liebig-Universität Gießen, Giessen, Germany* (2018).
- [Aye19] S. Ayet San Andrés *et al.* “High-resolution, accurate multiple-reflection time-of-flight mass spectrometry for short-lived, exotic nuclei of a few events in their ground and low-lying isomeric states.” *Physical Review C* **99** (2019) 064313.
- [Bab18] C. Babcock *et al.* “Mass measurements of neutron-rich indium isotopes toward the $N = 82$ shell closure.” *Physical Review C* **97** (2018) 024312.
- [Bal02] D. L. Balabanski, M. Ionescu-Bujor, A. Iordachescu *et al.* “Spin, quadrupole moment, and deformation of the magnetic-rotational band head in ^{193}Pb .” *Physical Review C* **83** (2002) 014304.
- [Bal19] J. Ballof *et al.* “The upgraded ISOLDE yield database - A new tool to predict beam intensities.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* (2019) (Online).
- [Bar13] A. E. Barzakh *et al.* “Changes in the mean-square charge radii and magnetic moments of neutron-deficient Tl isotopes.” *Physical Review C* **88** (2013) 024315.
- [Bar16] A. E. Barzakh *et al.* “Laser spectroscopy studies of intruder states in $^{193,195,197}\text{Bi}$.” *Physical Review C* **94** (2016) 024334.
- [Bar17a] A. E. Barzakh *et al.* “Changes in mean-squared charge radii and magnetic moments of $^{179-184}\text{Tl}$ measured by in-source laser spectroscopy.” *Physical Review C* **95** (2017) 014324.

- [Bar17b] A. E. Barzakh *et al.* “Onset of deformation in neutron-deficient Bi isotopes studied by laser spectroscopy.” *Physical Review C* **95** (2017) 044324.
- [Bar18] A. E. Barzakh *et al.* “Shell effect in the mean square charge radii and magnetic moments of bismuth isotopes near $N = 126$.” *Physical Review C* **97** (2018) 014322.
- [Bar19] A. E. Barzakh *et al.* “Inverse odd-even staggering in nuclear charge radii and possible octupole collectivity in $^{217,218,219}\text{At}$ revealed by in-source laser spectroscopy.” *Physical Review C* **99** (2019) 054317.
- [Bas07] B. Bastin *et al.* “Collapse of the $N = 28$ Shell Closure in ^{42}Si .” *Physical Review Letters* **99** (2007) 022503.
- [Bau73] R. Bauer, J. Speth *et al.* “Magnetic multipole moments and transition probabilities of single-particle states around ^{208}Pb .” *Nuclear Physics A* **209** (1973) 535–556.
- [Bec48] G. E. Becker and P. Kusch. “On the Hyperfine Structure of Gallium.” *Physical Review* **73** (1948) 584–591.
- [Bem73] C. E. Bemis Jr. *et al.* “X-Ray Identification of Element 104.” *Physical Review Letters* **31** (1973) 647–650.
- [Ben06] M. Bender, G. F. Bertsch and P. -H. Heenen. “Global study of quadrupole correlation effects.” *Physical Review C* **73** (2006) 034322.
- [Ben99] M. Bender *et al.* “Shell structure of superheavy nuclei in self-consistent mean-field models.” *Physical Review C* **60** (1999) 034304.
- [Ber09] J. S. Berryman, K. Minamisono, W. F. Rogers *et al.* “Doubly-magic nature of ^{56}Ni : Measurement of the ground state nuclear magnetic dipole moment of ^{55}Ni .” *Physical Review C* **79** (2009) 064305.
- [Ber85] D. Berdichevsky and F. Tondeur. “Nuclear core densities, isotope shifts, and the parameterization of the droplet model.” *Zeitschrift für Physik A - Atoms and Nuclei* **322** (1985) 141–147.
- [Bie52] L. C. Biedenharn, J. M. Blatt and M. E. Rose. “Some Properties of the Racah and Associated Coefficients.” *Reviews of Modern Physics* **24** (1952) 249–257.
- [Bir91] D. J. S. Birch *et al.* “The fluorescence properties of DCM.” *Chemical Physics Letters* **178** (1991) 177–184.
- [Bis16] M. L. Bissell *et al.* “Cu charge radii reveal a weak sub-shell effect at $N = 40$.” *Physical Review C* **93** (2016) 064318.
- [Bla13] K. Blaum, J. Dilling and W. Nörtershäuser. “Precision atomic physics techniques for nuclear physics with radioactive beams.” *Physica Scripta* T152 (2013) 014017.

- [Bli54] R. J. Blin-Stoyle and M. A. Perks. “The Deviations of Nuclear Magnetic Moments from the Schmidt Lines.” *Proceedings of the Physical Society: Section A* **67** (1954) 885–894.
- [Blu87] S. A. Blundell *et al.* “A reformulation of the theory of field isotope shift in atoms.” *Journal of Physics B: Atomic and Molecular Physics* **20** (1987) 3663–3681.
- [Boh50] A. Bohr and V. F. Weisskopf. “The Influence of Nuclear Structure on the Hyperfine Structure of Heavy Elements.” *Physical Review* **77** (1950) 94–98.
- [Boh98] A. Bohr and B. R. Mottelson. “Nuclear Structure, Volume I: Single-Particle Motion.” *World Scientific Publishing Company* (1998).
- [Bol92] G. Bollen *et al.* “Resolution of nuclear ground and isomeric states by a Penning trap mass spectrometer.” *Physical Review C* **46** (1992) R2140(R).
- [Bon04] S. L. Bondarev *et al.* “Fluorescence and Electronic Structure of the Laser Dye DCM in Solution and in Polymethylmethacrylate.” *Journal of Applied Spectroscopy* **71** (2004) 194–201.
- [Bor17] M. J. G. Borge and B. Jonson. “ISOLDE past, present and future.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 044011.
- [Bor87] W. Borchers *et al.* “Hyperfine structure and isotope shift investigations in $^{202-222}\text{Rn}$ for the study of nuclear structure beyond $Z = 82$.” *Hyperfine Interactions* **34** (1987) 25–29.
- [Bri94] D. M. Brink and G. R. Satchler. “Angular Momentum.” *Clarendon Press* 3rd Edition (1994).
- [Bro10] B. A. Brown and A. F. Lisetskiy. (Unpublished). (2010).
- [Bro87] B. A. Brown and B. H. Wildenthal. “Empirically optimum M1 operator for sd-shell nuclei.” *Nuclear Physics A* **474** (1987) 290–306.
- [Buc90] F. Buchinger *et al.* “Systematics of nuclear ground state properties in $^{78-100}\text{Sr}$ by laser spectroscopy.” *Physical Review C* **41** (1990) 2883–2897.
- [Bud14] I. Budinčević *et al.* “Laser spectroscopy of francium isotopes at the borders of the region of reflection asymmetry.” *Physical Review C* **90** (2014) 014317.
- [Bür05] A. Bürger *et al.* “Relativistic Coulomb excitation of neutron-rich $^{54,56,58}\text{Cr}$: On the pathway of magicity from $N = 40$ to $N = 32$.” *Physics Letters B* **622** (2005) 29–34.
- [Bur57] E. M. Burbidge *et al.* “Synthesis of the Elements in Stars.” *Reviews of Modern Physics* **29** (1957) 547–650.

- [Buu88] E. P. Buurman, P. H. M. Uylings and A. Dönszelmann. “On the intercombination line $4s^24p\ ^2P - 4s4p^2\ ^4P$ in the spectrum of neutral gallium.” *Zeitschrift für Physik D - Atoms, Molecules and Clusters* **8** (1988) 7–10.
- [Cal73] P. T. Callaghan, M. Kaplan and N. J. Stone. “The magnetic dipole moment of ^{55}Co .” *Nuclear Physics A* **201** (1973) 561–569.
- [Cam16] P. Campbell, I. D. Moore and M. R. Pearson. “Laser spectroscopy for nuclear structure physics.” *Progress in Particle and Nuclear Physics* **86** (2016) 127–180.
- [Car86] J. Carlsson *et al.* “Experimental and theoretical investigation of radiative lifetimes in neutral gallium.” *Zeitschrift für Physik D - Atoms, Molecules and Clusters* **3** (1986) 345–351.
- [Cas90] B. Castel and I. S. Towner. “Modern Theories of Nuclear Moments.” *Clarendon Press* 1st Edition (1990).
- [Cat03] R. Catherall *et al.* “Radioactive ion beams produced by neutron-induced fission at ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **204** (2003) 235–239.
- [Cat17] R. Catherall and others. “The ISOLDE facility.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 094002.
- [Che09] B. Cheal *et al.* “Laser Spectroscopy of Niobium Fission Fragments: First Use of Optical Pumping in an Ion Beam Cooler Buncher.” *Physical Review Letters* **102** (2009) 222501.
- [Che10a] B. Cheal *et al.* “Discovery of a long-lived low-lying isomeric state in ^{80}Ga .” *Physical Review C* **82** (2010) 051302(R).
- [Che10b] B. Cheal, E. Mané *et al.* “Nuclear Spins and Moments of Ga Isotopes Reveal Sudden Structural Changes between $N = 40$ and $N = 50$.” *Physical Review Letters* **104** (2010) 252502.
- [Che12a] B. Cheal *et al.* “Laser spectroscopy of gallium isotopes beyond $N = 50$.” *Journal of Physics: Conference Series* **381** (2012) 012071.
- [Che12b] B. Cheal, T. E. Cocolios and S. Fritzsche. “Laser spectroscopy of radioactive isotopes: Role and limitations of accurate isotope-shift calculations.” *Physical Review A* **86** (2012) 042501.
- [Coc09] T. E. Cocolios *et al.* “Magnetic Dipole Moment of $^{57,59}\text{Cu}$ Measured by In-Gas-Cell Laser Spectroscopy.” *Physical Review Letters* **103** (2009) 102501.
- [Coc10] T. E. Cocolios *et al.* “Structure of ^{191}Pb from α - and β -decay spectroscopy.” *Journal of Physics G: Nuclear and Particle Physics* **37** (2010) 125103.

- [Coc11] T. E. Cocolios, W. Dexters, M. D. Seliverstov *et al.* “Early Onset of Ground State Deformation in Neutron Deficient Polonium Isotopes.” *Physical Review Letters* **106** (2011) 052503.
- [Coc13a] T. E. Cocolios *et al.* “Study of the stability of the gallium isotopes beyond the $N = 50$ neutron shell closure.” *CERN-INTC P-375* (2013).
- [Coc13b] T. E. Cocolios *et al.* “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* **317** (2013) 565–569.
- [Coc16] T. E. Cocolios, R. P. de Groote *et al.* “High-resolution laser spectroscopy with the Collinear Resonance Ionisation Spectroscopy (CRIS) experiment at CERN-ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **376** (2016) 284–287.
- [Coc17] T. E. Cocolios. “A new perspective on charge radii around $Z = 82$.” *Hyperfine Interactions* **238** (2017) 16–23.
- [Coc85] A. Coc *et al.* “Hyperfine structures and isotope shifts of $^{207-213}, ^{220-228}\text{Fr}$; Possible evidence of octupolar deformation.” *Physics Letters B* **163** (1985) 66–70.
- [Col12a] ATLAS Collaboration. “Observation of a new particle in the search for the Standard Model Higgs boson with the ATLAS detector at the LHC.” *Physics Letters B* **716** (2012) 1–29.
- [Col12b] CMS Collaboration. “Observation of a new boson at a mass of 125 GeV with the CMS experiment at the LHC.” *Physics Letters B* **716** (2012) 30–61.
- [Col14] R. Collister *et al.* “Isotope shifts in francium isotopes $^{206-213}\text{Fr}$ and ^{221}Fr .” *Physical Review A* **90** (2014) 052502.
- [Con81] J. P. Connerade and M. A. Baig. “On the sp^2 configuration in the Ga I, In I, and Tl I isoelectronic sequences.” *Journal of Physics B: Atomic and Molecular Physics* **14** (1981) 29–38.
- [Coo19] B. S. Cooper *et al.* “A compact RFQ cooler buncher for CRIS experiments.” *Hyperfine Interactions* **240** (2019) 52–59.
- [Cor04] D. Cortina-Gil, J. Fernandez-Vasquez *et al.* “Shell Structure of the Near-Dripline Nucleus ^{23}O .” *Physical Review Letters* **93** (2004) 062501.
- [Cra49] M. F. Crawford and A. L. Schawlow. “Electron-Nuclear Potential Fields from Hyperfine Structure.” *Physical Review* **76** (1949) 1310–1317.

- [Cro68] P. D. Croft, J. M. Alexander and K. Street Jr. "Recoil Studies of Nuclear Reactions of Bi^{209} and Pb^{208} with O^{16} ." *Physical Review* **165** (1968) 1380–1390.
- [Cub18] J. G. Cubiss *et al.* "Charge radii and electromagnetic moments of $^{195-211}\text{At}$." *Physical Review C* **97** (2018) 054327.
- [Cub19] J. G. Cubiss, A. E. Barzakh, A. N. Andreyev *et al.* "Fine structure in the α decay of ^{218}At ." *Physical Review C* **99** (2019) 064317.
- [Cun53] J. G. Cuninghame. "The chain ^{79}As - ^{79m}Se in slow neutron fission of ^{235}U ." *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **44** (1953) 900–906.
- [Ćwi05] S. Ćwiok, P. -H. Heenen and W. Nazarewicz. "Shape coexistence and triaxiality in the superheavy nuclei." *Nature* **433** (2005) 705–709.
- [Cze13] M. Czerwiński *et al.* "Yrast excitation in the neutron-rich $N = 52$ isotones." *Physical Review C* **88** (2013) 044314.
- [Dal54] R. T. Daly Jr. and J. H. Holloway. "Nuclear Magnetic Octupole Moments of the Stable Gallium Isotopes." *Physical Review* **96** (1954) 539–540.
- [Day17] T. Day Goodacre. "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, The University of Manchester, Manchester, United Kingdom* (2017).
- [De 07] H. De Witte *et al.* "Nuclear Charge Radii of Neutron-Deficient Lead Isotopes Beyond $N = 104$ Midshell Investigated by In-Source Laser Spectroscopy." *Physical Review Letters* **98** (2007) 112502.
- [Del18] C. Delafosse *et al.* "Pseudospin Symmetry and Microscopic Origin of Shape Coexistence in the ^{78}Ni Region: A Hint from Lifetime Measurements." *Physical Review Letters* **121** (2018) 192502.
- [Dem15] W. Demtröder. "Laser Spectroscopy 2: Applications of Laser Spectroscopy." *Springer* 5th Edition (2015).
- [Did17] F. Didierjean *et al.* "Neutron effective single-particle energies above ^{78}Ni : A hint from lifetime measurements in the $N = 51$ isotones ^{85}Se and ^{87}Kr ." *Physical Review C* **96** (2017) 044320.
- [Din05] D. -C. Dinca, R. V. F. Janssens, A. Gade *et al.* "Reduced transition probabilities to the first 2^+ state in $^{52,54,56}\text{Ti}$ and development of shell closures at $N = 32,34$." *Physical Review C* **71** (2005) 041302(R).
- [Din87] U. Dinger *et al.* "Nuclear moments and change in the charge-radii of neutron deficient lead isotopes." *Zeitschrift für Physik A - Atomic Nuclei* **328** (1987) 253–254.

- [Dob94] J. Dobaczewski *et al.* “Nuclear shell structure at particle drip lines.” *Physical Review Letters* **72** (1994) 981–984.
- [Dra06] G. W. F. Drake. “Springer Handbook of Atomic, Molecular, and Optical Physics.” *Springer* 1st Edition (2006).
- [Dud19] J. Dudouet *et al.* “Excitations of the magic $N = 50$ neutron-core revealed in ^{81}Ga .” *Physical Review C* **100** (2019) 011301(R).
- [Duo87] H. T. Duong *et al.* “First Observation of the Blue Optical Lines of Francium.” *Europhysics Letters* **3** (1987) 175–182.
- [Dut91] S. B. Dutta *et al.* “Measurement of isotope shift and hyperfine splitting of $^{190,191,193,197}\text{Pb}$ isotopes by collinear laser spectroscopy.” *Zeitschrift für Physik A - Hadrons and Nuclei* **341** (1991) 39–45.
- [Dzu05] V. A. Dzuba, W. R. Johnson and M. S. Safronova. “Calculation of isotope shifts for cesium and francium.” *Physical Review A* **72** (2005) 022503.
- [Ebe87] J. Eberz *et al.* “Spins, moments and mean square charge radii of $^{104-127}\text{In}$ determined by laser spectroscopy.” *Nuclear Physics A* **464** (1987) 9–28.
- [Ebr16] J. -P. Ebran *et al.* “Spin-orbit interaction in relativistic nuclear structure models.” *Physical Review C* **94** (2016) 024304.
- [Eck30] C. Eckart. “The Application of Group theory to the Quantum Dynamics of Monatomic Systems.” *Reviews of Modern Physics* **2** (1930) 305–380.
- [Edm57] A. R. Edmonds. “Angular Momentum in Quantum Mechanics.” *Princeton University Press* 1st Edition (1957).
- [Ehl68] V. J. Ehlers *et al.* “Hyperfine Structure of ^{67}Ga and ^{72}Ga .” *Physical Review* **176** (1968) 25–42.
- [Eks08] A. Ekström *et al.* “ $0_{\text{gs}}^+ \rightarrow 2_1^+$ Transition Strengths in ^{106}Sn and ^{108}Sn .” *Physical Review Letters* **101** (2008) 012502.
- [Eks78] C. Ekström *et al.* “Nuclear Ground State Spins of the Francium Isotopes $^{208-213,220-222}\text{Fr}$.” *Physica Scripta* **18** (1978) 51–53.
- [Eti15] A. Etilé, D. Verney *et al.* “Low-lying intruder and tensor-driven structures in ^{82}As revealed by β decay at a new movable-tape-based experimental setup.” *Physical Review C* **91** (2015) 064317.
- [Ewa82] G. T. Ewan *et al.* “New isomeric states in ^{218}Fr and ^{214}At .” *Nuclear Physics A* **380** (1982) 423–437.
- [Far16] G. J. Farooq-Smith, T. E. Cocolios *et al.* “Laser and decay spectroscopy of the short-lived isotope ^{214}Fr in the vicinity of the $N = 126$ shell closure.” *Physical Review C* **94** (2016) 054305.

- [Far17] G. J. Farooq-Smith, A. R. Vernon *et al.* “Probing the ^{31}Ga ground-state properties in the region near $Z = 28$ with high-resolution laser spectroscopy.” *Physical Review C* **96** (2017) 044324.
- [Fay96] S. A. Fayans and D. Zawischa. “Towards a better parameterization of the nuclear pairing force: density dependence with gradient term.” *Physics Letters B* **383** (1996) 19–23.
- [Fer17] R. Ferrer *et al.* “Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion.” *Nature Communications* **8** (2017) 14520.
- [Fin15] D. A. Fink, T. E. Cocolios *et al.* “In-source Laser Spectroscopy with the Laser Ion Source and Trap: First Direct Study of the Ground-State Properties of $^{217,219}\text{Po}$.” *Physical Review X* **5** (2015) 011018.
- [Fla09] K. T. Flanagan, P. Vingerhoets *et al.* “Nuclear Spins and Magnetic Moments of $^{71,73,75}\text{Cu}$: Inversion of $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ Levels in ^{75}Cu .” *Physical Review Letters* **103** (2009) 142501.
- [Fla10] K. T. Flanagan, P. Vingerhoets *et al.* “Experimental determination of an $I^\pi = 2^-$ ground state in $^{72,74}\text{Cu}$.” *Physical Review C* **82** (2010) 041302(R).
- [Fla12] K. T. Flanagan *et al.* “Nuclear moments, charge radii and spins of the ground and isomeric states in ^{175}Yb and ^{177}Yb .” *Journal of Physics G: Nuclear and Particle Physics* **39** (2012) 125101.
- [Fla13a] K. T. Flanagan. “CRIS: A New Sensitive Device for Laser Spectroscopy of Exotic Nuclei.” *Nuclear Physics News* **23** (2013) 24–26.
- [Fla13b] K. T. Flanagan, K. M. Lynch *et al.* “Collinear Resonance Ionization Spectroscopy of Neutron-Deficient Francium Isotopes.” *Physical Review Letters* **111** (2013) 212501.
- [Flo52] B. H. Flowers. “Magnetic moments and quadrupole moments of odd-mass nuclei in jj-coupling.” *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **43** (1952) 1330–1334.
- [Fra01] S. Franchoo *et al.* “Monopole migration in $^{69,71,73}\text{Cu}$ observed from β decay of laser-ionized $^{68-74}\text{Ni}$.” *Physical Review C* **64** (2001) 054308.
- [Frå08] H. Frånberg, P. Delahaye *et al.* “Off-line commissioning of the ISOLDE cooler.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **266** (2008) 4502–4504.

- [Fra98] S. Franchoo *et al.* “Beta Decay of $^{68-74}\text{Ni}$ and Level Structure of Neutron-Rich Cu Isotopes.” *Physical Review Letters* **81** (1998) 3100–3103.
- [Fri04] G. Fricke and K. Heilig. “Nuclear Charge Radii.” *Springer-Verlag* (2004).
- [Fri12] S. Fritzsche. “The RATIP program for relativistic calculations of atomic transition, ionization and recombination properties.” *Computer Physics Communications* **183** (2012) 1525–1559.
- [Gaf13] L. P. Gaffney *et al.* “Studies of pear-shaped nuclei using accelerated radioactive beams.” *Nature* **497** (2013) 199–204.
- [Gai82] J. E. Gaiser. “Charmonium Spectroscopy from Radiative Decays of the J/ψ and ψ' .” *Ph.D. thesis, Stanford University, California, USA* (1982).
- [Gal12] A. T. Gallant *et al.* “New Precision Mass Measurements of Neutron-Rich Calcium and Potassium Isotopes and Three-Nucleon Forces.” *Physical Review Letters* **109** (2012) 032506.
- [Gar15a] R. F. Garcia Ruiz. “Investigating the possible magicity of $N = 32, 34$ in exotic Ca isotopes using laser spectroscopy methods.” *Ph.D. thesis, KU Leuven, Belgium* (2015).
- [Gar15b] R. F. Garcia Ruiz *et al.* “Ground-state electromagnetic moments of calcium isotopes.” *Physical Review C* **91** (2015) 041304(R).
- [Gar16] R. F. Garcia Ruiz *et al.* “Unexpectedly large charge radii of neutron-rich calcium isotopes.” *Nature Physics* **12** (2016) 594–598.
- [Gar18] R. F. Garcia Ruiz *et al.* “High-Precision Multiphoton Ionization of Accelerated Laser-Ablated Species.” *Physical Review X* **8** (2018) 041005.
- [Ger03] V. Gerginov, A. Derevianko and C. E. Tanner. “Observation of the Nuclear Magnetic Octupole Moment of ^{133}Cs .” *Physical Review Letters* **91** (2003) 072501.
- [Gin18] W. Gins, R. P. de Groote *et al.* “Analysis of counting data: Development of the SATLAS Python package.” *Computer Physics Communications* **222** (2018) 286–294.
- [Gin19] W. Gins. “Development of a dedicated laser-polarization beamline for ISOLDE-CERN.” *Ph.D. thesis, KU Leuven, Belgium* (2019).
- [God13] P. M. Goddard, P. D. Stevenson and A. Rios. “Charge Radius Isotope Shift Across the $N = 126$ Shell Gap.” *Physical Review Letters* **110** (2013) 032503.

- [Gor19] C. Gorges, L. V. Rodríguez *et al.* “Laser Spectroscopy of Neutron-Rich Tin Isotopes: A Discontinuity in Charge Radii across the $N = 82$ Shell Closure.” *Physical Review Letters* **122** (2019) 192502.
- [Got12] A. Gottardo *et al.* “New Isomers in the Full Seniority Scheme of Neutron-Rich Lead Isotopes: The Role of Effective Three-Body Forces.” *Physical Review Letters* **109** (2012) 162502.
- [Got14] A. Gottberg *et al.* “Experimental tests of an advanced proton-to-neutron converter at ISOLDE-CERN.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **336** (2014) 143–148.
- [Got16] A. Gottardo *et al.* “First Evidence of Shape Coexistence in the ^{78}Ni Region: Intruder 0_2^+ State in ^{80}Ge .” *Physical Review Letters* **116** (2016) 182501.
- [Gri08] G. F. Grinyer *et al.* “High-precision half-life determination for the superallowed β^+ emitter ^{62}Ga .” *Physical Review C* **77** (2008) 015501.
- [Gro15] R. P. de Groote, I. Budinčević *et al.* “Use of a Continuous Wave Laser and Pockels Cell for Sensitive High-Resolution Collinear Resonance Ionization Spectroscopy.” *Physical Review Letters* **115** (2015) 132501.
- [Gro17a] R. P. de Groote. “High resolution collinear resonance ionization spectroscopy of neutron-rich $^{76,77,78}\text{Cu}$ isotopes.” *Ph.D. thesis, KU Leuven, Belgium* (2017).
- [Gro17b] R. P. de Groote *et al.* “Dipole and quadrupole moments of $^{73-78}\text{Cu}$ as a test of the robustness of the $Z = 28$ shell closure near ^{78}Ni .” *Physical Review C* **96** (2017) 041302(R).
- [Gro17c] R. P. de Groote, K. M. Lynch, S. G. Wilkins *et al.* “Achieving sensitive, high-resolution laser spectroscopy at CRIS.” *Hyperfine Interactions* **238** (2017) 5–11.
- [Gro17d] R. P. de Groote, M. Verlinde, V. Sonnenschein *et al.* “Efficient, high-resolution resonance laser ionization spectroscopy using weak transitions to long-lived excited states.” *Physical Review A* **95** (2017) 032502.
- [Hag16] G. Hagen, G. R. Jansen and T. Papenbrock. “Structure of ^{78}Ni from First-Principles Computations.” *Physical Review Letters* **117** (2016) 172501.
- [Hah21] O. Hahn. “Über ein neues radioaktives Zerfallsprodukt im Uran.” *Naturwissenschaften* **9** (1921) 84.

- [Hak08] J. Hakala, S. Rahaman *et al.* “Evolution of the $N = 50$ Shell Gap Energy towards ^{78}Ni .” *Physical Review Letters* **101** (2008) 052502.
- [Ham13] H-W. Hammer, A. Nogga and A. Schwenk. “Colloquium : Three-body forces: From cold atoms to nuclei.” *Reviews of Modern Physics* **85** (2013) 197–217.
- [Han87] P. G. Hansen and B. Jonson. “The Neutron Halo of Extremely Neutron-Rich Nuclei.” *Europhysics Letters* **4** (1987) 409–414.
- [Har15] J. C. Hardy and I. S. Towner. “Superaligned $0^+ \rightarrow 0^+$ nuclear β decays: 2014 critical survey, with precise results for V_{ud} and CKM unitarity.” *Physical Review C* **91** (2015) 025501.
- [Hee02] P. -H. Heenen and W. Nazarewicz. “Quest for superheavy nuclei.” *Europhysics News* **33** (2002) 5–9.
- [Her10] A. Herlert. “The ISOLDE Facility.” *Nuclear Physics News* **20** (2010) 5–12.
- [Hey16] H. Heylen *et al.* “Changes in nuclear structure along the Mn isotopic chain studied via charge radii.” *Physical Review C* **94** (2016) 054321.
- [Hey90] K. L. G. Heyde. “The Nuclear Shell Model.” *Springer* 1st Edition (1990).
- [Hil92] Th. Hilberath *et al.* “Ground-state properties of neutron-deficient platinum isotopes.” *Zeitschrift für Physik A - Hadrons and Nuclei* **342** (1992) 1–15.
- [Hir17a] Y. Hirayama *et al.* “Ionization cross section, pressure shift and isotope shift measurements of osmium.” *Journal of Physics B: Atomic, Molecular and Optical Physics* **50** (2017) 215203.
- [Hir17b] Y. Hirayama, M. Mukai, Y. X. Watanabe *et al.* “In-gas-cell laser spectroscopy of the magnetic dipole moment of the $N \approx 126$ isotope ^{199}Pt .” *Physical Review C* **96** (2017) 014307.
- [Hjo95] M. Hjorth-Jensen, T. T. S. Kuo and E. Osnes. “Realistic effective interactions for nuclear systems.” *Physics Reports* **261** (1995) 125–270.
- [Hof09] C. R. Hoffman *et al.* “Evidence for a doubly magic ^{24}O .” *Physics Letters B* **672** (2009) 17–21.
- [Hof81] P. Hoff and B. Fogelberg. “Properties of strongly neutron-rich isotopes of germanium and arsenic.” *Nuclear Physics A* **368** (1981) 210–236.
- [Hon02a] H. Honma *et al.* “Effective interaction for pf -shell nuclei.” *Physical Review C* **65** (2002) 061301(R).

- [Hon02b] M. Honma *et al.* “Full pf-shell calculations with a new effective interaction.” *Nuclear Physics A* **704** (2002) 134–143.
- [Hon04] M. Honma *et al.* “New effective interaction for *pf*-shell nuclei and its implications for the stability of the $N = Z = 28$ closed core.” *Physical Review C* **69** (2004) 034335.
- [Hon05] H. Honma *et al.* “Shell-model description of neutron-rich pf-shell nuclei with a new effective interaction GXPF1.” *The European Physical Journal A - Hadrons and Nuclei* **25** (2005) 499–502.
- [Hon09] H. Honma *et al.* “New effective interaction for $f_5p_{g_9}$ -shell nuclei.” *Physical Review C* **80** (2009) 064323.
- [Hul06] M. Hult *et al.* “Underground search for the decay of $^{180}\text{Ta}^m$.” *Physical Review C* **74** (2006) 054311.
- [Jac79] P. Jacquinet and R. Klapisch. “Hyperfine spectroscopy of radioactive atoms.” *Reports on Progress in Physics* **42** (1979) 773–832.
- [Jan02] R. V. F. Janssens *et al.* “Structure of $^{52,54}\text{Ti}$ and shell closures in neutron-rich nuclei above ^{48}Ca .” *Physics Letters B* **546** (2002) 55–62.
- [Käl89] W. Kälber, J. Rink *et al.* “Nuclear Radii of Thorium Isotopes from Laser Spectroscopy of Stored Ions.” *Zeitschrift für Physik A - Atomic Nuclei* **334** (1989) 103–108.
- [Kam12] D. Kameda *et al.* “Observation of new microsecond isomers among fission products from in-flight fission of 345 MeV/nucleon ^{238}U .” *Physical Review C* **86** (2012) 054319.
- [Kan09] R. Kanungo, C. Nociforo, A. Prochazka *et al.* “One-Neutron Removal Measurement Reveals ^{24}O as a New Doubly Magic Nucleus.” *Physical Review Letters* **102** (2009) 152501.
- [Kei95] M. Keim *et al.* “Laser-spectroscopy measurements of $^{72-96}\text{Kr}$ spins, moments and charge radii.” *Nuclear Physics A* **586** (1995) 219–239.
- [Kes08] T. Kessler *et al.* “An injection-seeded high-repetition rate Ti:Sapphire laser for high-resolution spectroscopy and trace analysis of rare isotopes.” *Laser Physics* **18** (2008) 842–849.
- [Kin84] W. H. King. “Isotope Shifts in Atomic Spectra.” *Springer* 1st Edition (1984).
- [Kir18] O. S. Kirsebom *et al.* “First Accurate Normalization of the β -delayed α Decay of ^{16}N and Implications for the $^{12}\text{C}(\alpha, \gamma)^{16}\text{O}$ Astrophysical Reaction Rate.” *Physical Review Letters* **121** (2018) 142701.
- [Kno10] G. F. Knoll. “Radiation Detection and Measurement.” *John Wiley & Sons* 4th Edition (2010).

- [Köe05] U. Köester *et al.* “ISOLDE beams of neutron-rich zinc isotopes: yields, release, decay spectroscopy.” *AIP Conference Proceedings* **798** (2005) 315–322.
- [Koh13] M. Kohno. “Nuclear and neutron matter G -matrix calculations with a chiral effective field theory potential including effects of three-nucleon interactions.” *Physical Review C* **88** (2013) 064005.
- [Kol13] K. Kolos *et al.* “Probing nuclear structures in the vicinity of ^{78}Ni with β - and βn -decay spectroscopy of ^{84}Ga .” *Physical Review C* **88** (2013) 047301.
- [Kor13] A. Korgul *et al.* “Experimental study of the β - γ and β - $n\gamma$ decay of the neutron-rich nucleus ^{85}Ga .” *Physical Review C* **88** (2013) 044330.
- [Kor16] A. Korgul, K. P. Rykaczewski, R. K. Grzywacz *et al.* “ β and β - n decay of the neutron-rich ^{84}Ge nucleus.” *Physical Review C* **93** (2016) 064324.
- [Kös00a] U. Köster. “Yields and spectroscopy of radioactive isotopes at LOHENGRIN and ISOLDE.” *Ph.D. thesis, Technischen Universität München, Munich, Germany* (2000).
- [Kös00b] U. Köster *et al.* “Isomer separation and measurement of nuclear moments with the ISOLDE RILIS.” *Hyperfine Interactions* **127** (2000) 417–420.
- [Kös03] U. Köster *et al.* “Oxide fiber targets at ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **204** (2003) 303–313.
- [Kös11] U. Köster *et al.* “In-source laser spectroscopy of $^{75,77,78}\text{Cu}$: Direct evidence for a change in the quasiparticle energy sequence in $^{75,77}\text{Cu}$ and an absence of longer-lived isomers in ^{78}Cu .” *Physical Review C* **84** (2011) 034320.
- [Kos19a] Á Koszorús *et al.* “Resonance ionization schemes for high resolution and high efficiency studies of exotic nuclei at the CRIS experiment.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* (2019) (Online).
- [Kos19b] Á Koszorús, X. F. Yang *et al.* “Precision measurements of the charge radii of potassium isotopes.” *Physical Review C* **100** (2019) 034304.
- [Kow91] D. Kowalewska *et al.* “Isotope shifts and hyperfine structure in polonium isotopes by atomic-beam laser spectroscopy.” *Physical Review A* **44** (1991) R1442(R).

- [Kre13] S. Kreim *et al.* “Recent exploits of the ISOLTRAP mass spectrometer.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **317** (2013) 492–500.
- [Krö88] U. Krönert *et al.* “Observation of strongly deformed ground-state configurations in ^{184}Au and ^{183}Au by laser spectroscopy.” *Zeitschrift für Physik A - Atomic Nuclei* **331** (1988) 521–522.
- [Kur03] J. Kurpeta, A. Plochoki, A. N. Andreyev *et al.* “Isomeric and ground-state decay of ^{215}Bi .” *The European Physical Journal A - Hadrons and Nuclei* **18** (2003) 31–37.
- [Laa16] M. Laatiaoui *et al.* “Atom-at-a-time laser resonance ionization spectroscopy of nobelium.” *Nature* **538** (2016) 495–498.
- [Le 99] F. Le Blanc *et al.* “Large odd-even radius staggering in the very light platinum isotopes from laser spectroscopy.” *Physical Review C* **60** (1999) 054310.
- [Leb09] M. Lebois *et al.* “Experimental study of ^{84}Ga β decay: Evidence for a rapid onset of collectivity in the vicinity of ^{78}Ni .” *Physical Review C* **80** (2009) 044308.
- [Lép05] A. Lépine-Szily *et al.* “Anomalous behaviour of matter radii of proton-rich Ga, Ge, As, Se and Br nuclei.” *The European Physical Journal A - Hadrons and Nuclei* **25** (2005) 227–230.
- [Lia92] C. F. Liang, P. Paris. and R. K. Sheline. “Reflection asymmetry in the transitional odd-odd nucleus ^{220}Fr .” *International Journal of Modern Physics E* **1** (1992) 363–377.
- [Lic14] R. Lică *et al.* “Low-lying isomeric states in ^{80}Ga from the β^- Decay of ^{80}Zn .” *Physical Review C* **90** (2014) 014320.
- [Lie96] P. Lievens *et al.* “On the odd-even staggering of mean-square charge radii in the light krypton and strontium region.” *Europhysics Letters* **33** (1996) 11–16.
- [Lin77] C. G. Lindén. “A 480 ms isomeric $29/2^-$ -state of the $(p_{1/2}^{-2})_{0+} f_{5/2}^{-1}(i_{13/2}^{-2})_{12+}$ configuration in ^{203}Pb .” *Zeitschrift für Physik A - Atoms and Nuclei* **280** (1977) 51–60.
- [Lin81] A. Lindgård *et al.* “A beam-foil study of lifetimes and level excitations in gallium and thallium.” *Zeitschrift für Physik A - Atoms and Nuclei* **301** (1981) 1–10.
- [Liu19] H. N. Liu *et al.* “How Robust is the $N = 34$ Subshell Closure? First Spectroscopy of ^{52}Ar .” *Physical Review Letters* **122** (2019) 072502.

- [Lom02] B. Lommel *et al.* “Preparation of self-supporting carbon thin films.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **480** (2002) 199–203.
- [Lön82] T. Lönnroth, V. Rahkonen and B. Fant. “The low-seniority shell-model structure of ^{212}At .” *Nuclear Physics A* **376** (1982) 29–44.
- [Lui12] R. Luis *et al.* “Optimization studies of the CERN-ISOLDE neutron converter and fission target system.” *The European Physical Journal A - Hadrons and Nuclei* **48** (2012) 90–100.
- [Lun17] D. Lunney (on behalf of the ISOLTRAP Collaboration). “Extending and refining the nuclear mass surface with ISOLTRAP.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 064008.
- [Lyn13] K. M. Lynch. “Laser assisted nuclear decay spectroscopy: A new method for studying neutron-deficient francium.” *Ph.D. thesis, The University of Manchester, Manchester, United Kingdom* (2013).
- [Lyn14] K. M. Lynch *et al.* “Decay-Assisted Laser Spectroscopy of Neutron-Deficient Francium.” *Physical Review X* **4** (2014) 011055.
- [Lyn16] K. M. Lynch, T. E. Cocolios *et al.* “Combined high-resolution laser spectroscopy and nuclear decay spectroscopy for the study of the low-lying states in ^{206}Fr , ^{202}At , and ^{198}Bi .” *Physical Review C* **93** (2016) 014319.
- [Lyn17] K. M. Lynch, T. E. Cocolios *et al.* “A simple decay-spectroscopy station at CRIS-ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **844** (2017) 14–18.
- [Lyn18] K. M. Lynch, S. G. Wilkins *et al.* “Laser-spectroscopy studies of the nuclear structure of neutron-rich radium.” *Physical Review C* **97** (2018) 024309.
- [Mad16] M. Madurga *et al.* “Evidence for Gamow-Teller Decay of ^{78}Ni Core from Beta-Delayed Neutron Emission Studies.” *Physical Review Letters* **117** (2016) 092502.
- [Man09a] E. B. Mané Júnior. “High resolution laser spectroscopy of radioactive isotopes using a RFQ cooler-buncher at CERN-ISOLDE.” *Ph.D. thesis, The University of Manchester, Manchester, United Kingdom* (2009).
- [Man09b] E. Mané *et al.* “An ion cooler-buncher for high-sensitivity collinear laser spectroscopy at ISOLDE.” *The European Physical Journal A - Hadrons and Nuclei* **42** (2009) 503–507.

- [Man11a] E. Mané, B. Cheal *et al.* “Ground-state spins and moments of $^{72,74,76,78}\text{Ga}$ nuclei.” *Physical Review C* **84** (2011) 024303.
- [Man11b] E. Mané, A. Voss *et al.* “First Experimental Determination of the Charge Radius of ^{74}Rb and Its Application in Tests of the Unitarity of the Cabibbo-Kobayashi-Maskawa Matrix.” *Physical Review Letters* **107** (2011) 212502.
- [Mar11] K. Marinova, W. Geithner, M. Kowalska *et al.* “Charge radii of neon isotopes across the *sd* neutron shell.” *Physical Review C* **84** (2011) 034313.
- [Mar13] B. A. Marsh *et al.* “New developments of the in-source spectroscopy method at RILIS/ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **317** (2013) 550–556.
- [Mar18a] B. A. Marsh *et al.* “Characterization of the shape-staggering effect in mercury nuclei.” *Nature Physics* **14** (2018) 1163–1167.
- [Mar18b] Y. Martinez Palenzuela, B. A. Marsh *et al.* “Enhancing the extraction of laser-ionized beams from an arc discharge ion source volume.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **431** (2018) 59–66.
- [Mar19] Y. Martinez Palenzuela. “Characterization and optimization of a versatile laser and electron-impact ion source for radioactive ion beam production at ISOLDE and MEDICIS.” *Ph.D. thesis, KU Leuven, Belgium* (2019).
- [Mat15] T. Materna *et al.* “Low spin structure of ^{86}Se : Confirmation of γ collectivity at $N = 52$.” *Physical Review C* **92** (2015) 034305.
- [Mat16] V. Matoušek, M. Sedlák, M. Venhart *et al.* “TATRA: a versatile high-vacuum tape transportation system for decay studies at radioactive-ion beam facilities.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **812** (2016) 118–121.
- [May49] M. G. Mayer. “On Closed Shells in Nuclei. II.” *Physical Review* **75** (1949) 1969–1970.
- [Mic18] S. Michimasa *et al.* “Magic Nature of Neutrons in ^{54}Ca : First Mass Measurements of $^{55-57}\text{Ca}$.” *Physical Review Letters* **121** (2018) 022506.
- [Mil17] A. J. Miller, K. Minamisono, D. M. Rossi *et al.* “First determination of ground state electromagnetic moments of ^{53}Fe .” *Physical Review C* **96** (2017) 054314.

- [Mil19] A. J. Miller, K. Minamisono, A. Klose *et al.* “Proton superfluidity and charge radii in proton-rich calcium isotopes.” *Nature Physics* **15** (2019) 432–436.
- [Min08] K. Minamisono, P. F. Mantica *et al.* “Quadrupole moment of ^{37}K .” *Physics Letters B* **662** (2008) 389–395.
- [Min16] K. Minamisono, D. M. Rossi *et al.* “Charge Radii of Neutron Deficient $^{52,53}\text{Fe}$ Produced by Projectile Fragmentation.” *Physical Review Letters* **117** (2016) 252501.
- [Miy51] H. Miyazawa. “Deviations of Nuclear Magnetic Moments from the Schmidt Lines.” *Progress of Theoretical Physics* **6** (1951) 801–814.
- [Moo02] R. Moore, A. M. Bruce, P. Dendooven *et al.* “Character of an 8^- isomer of ^{130}Ba .” *Physics Letters B* **547** (2002) 200–204.
- [Mou18] M. Mougeot *et al.* “Precision Mass Measurements of $^{58-63}\text{Cr}$: Nuclear Collectivity Towards the $N = 40$ Island of Inversion.” *Physical Review Letters* **120** (2018) 232501.
- [Muk17] S. Mukhopadhyay, B. P. Crider and B. A. Brown. “Nuclear structure of ^{76}Ge from inelastic neutron scattering measurements and shell model calculations.” *Physical Review C* **95** (2017) 014327.
- [Mye74] W. D. Myers and W. J. Swiatecki. “The nuclear droplet model for arbitrary shapes.” *Annals of Physics* **84** (1974) 186–210.
- [Mye83] W. D. Myers and K. -H. Schmidt. “An update on droplet-model charge distributions.” *Nuclear Physics A* **410** (1983) 61–73.
- [Nak15] H. Nakada and T. Inakura. “Effects of three-nucleon spin-orbit interaction on isotope shifts of Pb nuclei.” *Physical Review C* **91** (2015) 021302(R).
- [Nav00] A. Navin *et al.* “Direct Evidence for the Breakdown of the $N = 8$ Shell Closure in ^{12}Be .” *Physical Review Letters* **85** (2000) 266–269.
- [Neu17] R. Neugart *et al.* “Collinear laser spectroscopy at ISOLDE: new methods and highlights.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 064002.
- [Ney03] G. Neyens. “Nuclear magnetic and quadrupole moments for nuclear structure research on exotic nuclei.” *Reports on Progress in Physics* **66** (2003) 633–689.
- [Nie12] R. Niembro *et al.* “Isotopic dependence of the nuclear charge radii and binding energies in the relativistic Hartree-Fock formalism.” *Physics of Atomic Nuclei* **75** (2012) 269–284.
- [NIS18] NIST.gov. “Atomic Spectra Database.” *National Institute of Standards and Technology* (2018).

- [NND18] NNDC.bnl.gov. “Evaluated Nuclear Structure Data File: Search and Retrieval.” *National Nuclear Data Center* (2018).
- [Nob18] NobelPrize.org. “The Nobel Prize in Physics 1963”. *Nobel Media AB* (2018).
- [Nör14] W. Nörtershäuser and C. Geppert. “Nuclear Charge Radii of Light Elements and Recent Developments in Collinear Laser Spectroscopy.” *Lecture Notes in Physics* 879 (2014) 233–292.
- [Oht96] T. Ohtsubo *et al.* “Measurement of the nuclear magnetic moments of ^{57}Ni and ^{59}Fe .” *Physical Review C* **54** (1996) 554–558.
- [Oli17] L. Olivier *et al.* “Persistence of the $Z = 28$ Shell Gap Around ^{78}Ni : First Spectroscopy of ^{79}Cu .” *Physical Review Letters* **119** (2017) 192501.
- [Oli77] J. J. Olivero and R. L. Longbothum. “Empirical fits to the Voigt line width: A brief review.” *Journal of Quantitative Spectroscopy and Radiative Transfer* **17** (1977) 233–236.
- [Ore80] M. J. Oreglia. “A Study of the Reactions $\psi' \rightarrow \gamma\gamma\psi$.” *Ph.D. thesis, Stanford University, California, USA* (1980).
- [Orl15] R. Orlandi *et al.* “Single-neutron orbits near ^{78}Ni : spectroscopy of the $N = 49$ isotope ^{79}Zn .” *Physics Letters B* 740 (2015) 298–302.
- [Ots01] T. Otsuka *et al.* “Magic Numbers in Exotic Nuclei and Spin-Isospin Properties of the NN Interaction.” *Physical Review Letters* **87** (2001) 082502.
- [Ots05] T. Otsuka *et al.* “Evolution of Nuclear Shells due to the Tensor Force.” *Physical Review Letters* **95** (2005) 232502.
- [Ots10] T. Otsuka *et al.* “Novel Features of Nuclear Forces and Shell Evolution in Exotic Nuclei.” *Physical Review Letters* **104** (2010) 012501.
- [Oza00] A. Ozawa *et al.* “New Magic Number, $N = 16$, near the Neutron Drip Line.” *Physical Review Letters* **84** (2000) 5493–5495.
- [Paa79] V. Paar. “A parabolic rule for the energy dependence on $x = I(I + 1)$ for proton-neutron multiplets in odd-odd nuclei.” *Nuclear Physics A* **331** (1979) 16–28.
- [Pap15] J. Papuga. “Structure of potassium isotopes studied with collinear laser spectroscopy.” *Ph.D. thesis, KU Leuven, Belgium* (2015).
- [Pas94] G. Passler, J. Rikovska *et al.* “Quadrupole moments and nuclear shapes of neutron-deficient gold isotopes.” *Nuclear Physics A* **580** (1994) 173–212.

- [Pea00] M. R. Pearson, P. Campbell, K. Leerungnavarat *et al.* “Nuclear moments and charge radii of bismuth isotopes.” *Journal of Physics G: Nuclear and Particle Physics* **26** (2000) 1829–1848.
- [Per06] O. Perru, D. Verney, F. Ibrahim *et al.* “Study of the $N = 50$ major shell effect toward ^{78}Ni at PARRNe: Evidence of a weak-coupling structure in $^{83}_{32}\text{Ge}_{51}$.” *The European Physical Journal A - Hadrons and Nuclei* **28** (2006) 307–312.
- [Per13] J. R. Persson. “Table of hyperfine anomaly in atomic systems.” *Atomic Data and Nuclear Data Tables* **99** (2013) 62–68.
- [Pie18] M. Piersa *et al.* “Investigation of Low-Lying States in ^{133}Sn Populated in the β Decay of ^{133}In Using Isomer-Selective Laser Ionization.” *Acta Physica Polonica B* **49** (2018) 523–528.
- [Pie19] M. Piersa *et al.* “ β decay of ^{133}In : γ emission from neutron-unbound states in ^{133}Sn .” *Physical Review C* **99** (2019) 024304.
- [Pol87] A. R. Poletti *et al.* “The low-lying yrast structure of ^{212}Po .” *Nuclear Physics A* **473** (1987) 595–604.
- [Pri01] J. I. Prisciandaro, P. F. Mantica, B. A. Brown *et al.* “New evidence for a subshell gap at $N = 32$.” *Physics Letters B* **510** (2001) 17–23.
- [Pro12] T. J. Procter *et al.* “Nuclear mean-square charge radii of $^{63,64,66,68-82}\text{Ga}$ nuclei: No anomalous behavior at $N = 32$.” *Physical Review C* **86** (2012) 034329.
- [Pro13] T. J. Procter. “New techniques of laser spectroscopy on exotic isotopes of gallium and francium.” *Ph.D. thesis, The University of Manchester, Manchester, United Kingdom* (2013).
- [Rac43] G. Racah. “Theory of Complex Spectra. III.” *Physical Review* **63** (1943) 367–382.
- [Raj13] M. M. Rajabali, K. M. Lynch and T. E. Cocolios. “A dedicated decay-spectroscopy station for the collinear resonance ionization experiment at ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **707** (2013) 35–39.
- [Ram19a] J. P. Ramos *et al.* “Design and tests for the new CERN-ISOLDE spallation source: an integrated tungsten converter surrounded by an annular UC_x target operated at 2000 °C.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* (2019) (Online).
- [Ram19b] João P. Ramos. “Private communication.” (2019).
- [Ree71] S. B. Reehal and R. A. Sorensen. “Odd-even staggering of nuclear radii.” *Nuclear Physics A* **161** (1971) 385–400.

- [Rei17] P.-G. Reinhart and W. Nazarewicz. “Toward a global description of nuclear charge radii: Exploring the Fayans energy density functional.” *Physical Review C* **95** (2017) 064328.
- [Rei19] M. P. Reiter *et al.* “Mass Measurements of Neutron-Rich Gallium Isotopes Refine Production of Nuclei of the First r -Process Abundance Peak During GW170817.” *arXiv:1810.11561v1* (2019).
- [Rei95] P. G. Reinhard and H. Flocard. “Nuclear effective forces and isotope shifts.” *Nuclear Physics A* **584** (1995) 467–488.
- [Ros32] J. E. Rosenthal and G. Breit. “The Isotope Shift in Hyperfine Structure.” *Physical Review* **41** (1932) 459–470.
- [Rot13] S. Rothe *et al.* “Narrow linewidth operation of the RILIS titanium: Sapphire laser at ISOLDE/CERN.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **317** (2013) 561–564.
- [Rut14] Sir E. Rutherford F.R.S. “The structure of the atom.” *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **27** (1914) 488–498.
- [Ryt91] A. Rytz. “Recommended energy and intensity values of alpha particles from radioactive decay.” *Atomic Data and Nuclear Data Tables* **47** (1991) 205–239.
- [Saf06] U. I. Safronova, T. E. Cowan and M. S. Safronova. “Excitation energies, hyperfine constants, E1 transition rates and lifetimes of $4s^2nl$ states in neutral gallium.” *Journal of Physics B: Atomic, Molecular and Optical Physics* **39** (2006) 749–764.
- [Saf18] M. S. Safronova *et al.* “Nuclear Charge Radii of ^{229}Th from Isotope and Isomer Shifts.” *Physical Review Letters* **121** (2018) 213001.
- [Sah15] B. K. Sahoo *et al.* “Correlation trends in the hyperfine structures of $^{210,212}\text{Fr}$.” *Physical Review A* **91** (2015) 042507.
- [San07] J. E. Sansonetti. “Spectroscopic Data for Neutral Francium (Fr I).” *Journal of Physical and Chemical Reference Data* **36** (2007) 497–507.
- [San09] S. Sanguinetti *et al.* “Accurate measurements of transition frequencies and isotope shifts of laser-trapped francium.” *Optics Letters* **34** (2009) 893–895.
- [San14] R. M. dos Santos Augusto *et al.* “CERN-MEDICIS (Medical Isotopes Collected from ISOLDE): A New Facility.” *Applied Sciences* **4** (2014) 265–281.
- [Sav90] G. Savard *et al.* “Laser spectroscopy of laser-desorbed gold isotopes.” *Nuclear Physics A* **512** (1990) 241–252.

- [Sch10] F. Schwellnus *et al.* “The laser ion source trap for highest isobaric selectivity in online exotic isotope production.” *Review of Scientific Instruments* **81** (2010) 02A515.
- [Sch18] S. Schmidt *et al.* “The nuclear magnetic moment of ^{208}Bi and its relevance for a test of bound-state strong-field QED.” *Physics Letters B* **779** (2018) 324–330.
- [Sch84] N. Schulz, S. Khazrouni *et al.* “High-spin states in ^{215}Fr .” *Journal of Physics G: Nuclear Physics* **10** (1984) 1201–1218.
- [Sel13] M. D. Seliverstov, T. E. Cocolios, W. Dexters *et al.* “Charge radii of odd- A $^{191-211}\text{Po}$ isotopes.” *Physics Letters B* **719** (2013) 362–366.
- [Sel14] M. D. Seliverstov, T. E. Cocolios, W. Dexters *et al.* “Electromagnetic moments of odd- A $^{193-203,211}\text{Po}$ isotopes.” *Physical Review C* **89** (2014) 034323.
- [Sel18] S. Sels. “Laser spectroscopy of neutron-deficient mercury isotopes and commissioning of a gas-jet based RFQ ion guide.” *Ph.D. thesis, KU Leuven, Belgium* (2018).
- [Sel19] S. Sels *et al.* “Shape staggering of midshell mercury isotopes from in-source laser spectroscopy compared with density-functional-theory and Monte Carlo shell-model calculations.” *Physical Review C* **99** (2019) 044306.
- [Seu56] H. E. Seuss and H. C. Urey. “Abundances of the Elements.” *Reviews of Modern Physics* **28** (1956) 53–74.
- [Sev17] N. Severijns and B. Blank. “Weak interaction physics at ISOLDE.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 074002.
- [Sha17] C. M. Shand *et al.* “Shell evolution beyond $Z = 28$ and $N = 50$: Spectroscopy of $^{81,82,83,84}\text{Zn}$.” *Physics Letters B* **773** (2017) 492–497.
- [Sha95] M. M. Sharma *et al.* “Isospin Dependence of the Spin-Orbit Force and Effective Nuclear Potentials.” *Physical Review Letters* **74** (1995) 3744–3747.
- [She88] R. K. Sheline. “Octupole deformation in odd-odd nuclei.” *Physical Review C* **37** (1988) 423(R).
- [She89] R. K. Sheline, R. L. Ponting *et al.* “Spectroscopy of the high-lying configurations in ^{210}Bi .” *Czechoslovak Journal of Physics B* **39** (1989) 22–54.
- [Shi07] T. Shirai *et al.* “Spectral Data for Gallium: Ga I through Ga XXXI.” *Journal of Physical and Chemical Reference Data* **36** (2007) 509–615.

- [Shi13] N. Shimizu. "Nuclear shell-model code for massive parallel computation, "KSHELL". " *arXiv:1310.5431v1* (2013).
- [Shi84] G. Shimkaveg *et al.* "Laser-Induced Nuclear Orientation of $1-\mu\text{s}$ $^{85}\text{Rb}^m$." *Physical Review Letters* **53** (1984) 2230–2233.
- [Sie09] K. Sieja, F. Nowacki *et al.* "Shell model description of zirconium isotopes." *Physical Review C* **79** (2009) 064310.
- [Sie13] K. Sieja, T. R. Rodríguez *et al.* "Laboratory versus intrinsic description of nonaxial nuclei above doubly magic ^{78}Ni ." *Physical Review C* **88** (2013) 034327.
- [Sim17] E. C. Simpson and M. Shelley. "Nuclear cartography: patterns in binding energies and subatomic structure." *Physics Education* **52** (2017) 064002.
- [Sin13] A. K. Singh, D. Angom and V. Natarajan. "Observation of the nuclear magnetic octupole moment of ^{173}Yb from precise measurements of the hyperfine structure in the $^3P_2$ state." *Physical Review A* **87** (2013) 012512.
- [Skw86] T. Skwarnicki. "A study of the radiative CASCADE transitions between the Upsilon-Prime and Upsilon resonances." *Ph.D. thesis, Institute of Nuclear Physics, Cracow, Poland* (1986).
- [Son17] V. Sonnenschein *et al.* "Characterization of a pulsed injection-locked Ti:sapphire laser and its application to high resolution resonance ionization spectroscopy of copper." *Laser Physics* **27** (2017) 085701.
- [Sor02] O. Sorlin *et al.* " $^{68}_{28}\text{Ni}_{40}$: Magicity versus Superfluidity." *Physical Review Letters* **88** (2002) 092501.
- [Sor08] O. Sorlin and M. -G. Porquet. "Nuclear magic numbers: New features far from stability." *Progress in Particle and Nuclear Physics* **61** (2008) 602–673.
- [Sót19] Z. Sóti, J. Magill and R. Dreher. "Karlsruhe Nuclide Chart - New 10th edition 2018." *EPJ Nuclear Science and Technologies* **5** (2019) 6.
- [Sri11] P. C. Srivastava. "Structure of $^{71-78}\text{Ga}$ isotopes in the $f_{5/2}pg_{9/2}$ and $fp_{9/2}$ spaces." *Journal of Physics G: Nuclear and Particle Physics* **39** (2011) 015102.
- [Sta08] A. L. Stancik and E. B. Brauns. "A simple asymmetric lineshape for fitting infrared absorption spectra." *Vibrational Spectroscopy* **47** (2008) 66–69.
- [Sta13] J. Stanja, Ch. Borgmann *et al.* "Mass spectrometry and decay spectroscopy of isomers across the $Z = 82$ shell closure." *Physical Review C* **88** (2013) 054304.

- [Ste07] I. Stefanescu, G. Georgiev *et al.* “Coulomb Excitation of $^{68,70}\text{Cu}$: First use of Postaccelerated Isomeric Beams.” *Physical Review Letters* **98** (2007) 122701.
- [Ste13] D. Steppenbeck *et al.* “Evidence for a new nuclear ‘magic number’ from the level structure of ^{54}Ca .” *Nature* **502** (2013) 207–210.
- [Ste15] D. Steppenbeck *et al.* “Low-Lying Structure of ^{50}Ar and the $N = 32$ Subshell Closure.” *Physical Review Letters* **114** (2015) 252501.
- [Ste17] Christine Steenkamp. “Private communication.” (2017).
- [Ste80] A. Steudel, U. Triebe and D. Wendlandt. “Isotope shift in Ni I and changes in mean-square nuclear charge radii of the stable Ni isotopes.” *Zeitschrift für Physik A - Atoms and Nuclei* **296** (1980) 189–193.
- [Sun93] D. Sundholm and J. Olsen. “Finite element multiconfiguration Hartree-Fock determination of the nuclear quadrupole moments of chlorine, potassium, and calcium isotopes.” *The Journal of Chemical Physics* **98** (1993) 7152–7158.
- [Taj93] N. Tajima *et al.* “Self-consistent calculation of charge radii of Pb isotopes.” *Nuclear Physics A* **551** (1993) 434–450.
- [Tan19] R. Taniuchi *et al.* “ ^{78}Ni revealed as a doubly magic stronghold against nuclear deformation.” *Nature* **569** (2019) 53–58.
- [Tei17] A. Teigelhöfer. “Isotope shift and hyperfine structure measurements on silver, actinium and astatine by in-source resonant ionization laser spectroscopy.” *Ph.D. thesis, The University of Manitoba, Winnipeg, Canada* (2017).
- [Thi18] J. Thielking *et al.* “Laser spectroscopic characterization of the nuclear-clock isomer $^{229\text{m}}\text{Th}$.” *Nature* **556** (2018) 321–325.
- [Thi81] C. Thibault *et al.* “Hyperfine structure and isotope shift of the D_2 line of $^{76-98}\text{Rb}$ and some of their isomers.” *Physical Review C* **23** (1981) 2720–2729.
- [Tor68] D. F. Torgerson, R. A. Gough and R. D. Macfarlane. “Alpha Decay of the Isomers of ^{214}Fr .” *Physical Review* **174** (1968) 1494–1499.
- [Tor70] D. F. Torgerson and R. D. Macfarlane. “Alpha-Decay studies of the $N = 127$ Isotones: ^{214}Fr , ^{215}Ra , and ^{216}Ac .” *Physical Review C* **2** (1970) 2309–2318.
- [Ulm86] G. Ulm *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 - Atomic Nuclei* **325** (1986) 247–259.

- [Vaj18] Zs. Vajita *et al.* “Proton single particle energies next to ^{78}Ni : Spectroscopy of ^{77}Cu via single proton knock-out reaction.” *Physics Letters B* **782** (2018) 99–103.
- [Van02] J. Van Roosbroeck. “Systematic nuclear-structure study of even-mass Zn and Cu isotopes between $N=40$ and 50.” *Ph.D. thesis, KU Leuven, Belgium* (2002).
- [Van04] J. Van Roosbroeck, C. Guénaut *et al.* “Unambiguous Identification of Three β -Decaying Isomers in ^{70}Cu .” *Physical Review Letters* **92** (2004) 112501.
- [Ven17a] M. Venhart, J. L. Wood *et al.* “Application of the Broad Energy Germanium detector: A technique for elucidating β -decay schemes which involve daughter nuclei with very low energy excited states.” *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **849** (2017) 112–118.
- [Ven17b] M. Venhart, J. L. Wood, M. Sedláč *et al.* “New systematic features in the neutron-deficient Au isotopes.” *Journal of Physics G: Nuclear and Particle Physics* **44** (2017) 074003.
- [Ver06] D. Verney *et al.* “Deformation change in light iridium nuclei from laser spectroscopy.” *The European Physical Journal A - Hadrons and Nuclei* **30** (2006) 489–518.
- [Ver07] D. Verney, F. Ibrahim *et al.* “Low-energy states of $^{81}_{31}\text{Ga}_{50}$: Proton structure of the nuclei close to ^{78}Ni .” *Physical Review C* **76** (2007) 054312.
- [Ver13] D. Verney *et al.* “Structure of ^{80}Ge revealed by the β decay of isomeric states in ^{80}Ga : Triaxiality in the vicinity of ^{78}Ni .” *Physical Review C* **87** (2013) 054307.
- [Ver17] D. Verney *et al.* “Pygmy Gamow-Teller resonance in the $N = 50$ region: New evidence from staggering of β -delayed neutron-emission probabilities.” *Physical Review C* **95** (2017) 054320.
- [Ver19a] A. R. Vernon *et al.* “Simulation of the relative atomic populations of elements $1 \leq Z \leq 89$ following charge exchange tested with collinear resonance ionization spectroscopy of indium.” *Spectrochimica Acta Part B: Atomic Spectroscopy* **153** (2019) 61–83.
- [Ver19b] A. R. Vernon, R. P. de Groote *et al.* “Optimising the Collinear Resonance Ionisation Spectroscopy (CRIS) experiment at CERN-ISOLDE.” *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* (2019) (Online).

- [Ver19c] E. Verstraelen, A. Teigelhöfer, W. Ryssens *et al.* “Search for octupole-deformed actinium isotopes using resonance ionization spectroscopy.” *Physical Review C* **100** (2019) 044321.
- [Vin10] P. Vingerhoets, K. T. Flanagan *et al.* “Nuclear spins, magnetic moments, and quadrupole moments of Cu isotopes from $N = 28$ to $N = 46$: Probes for core polarization effects.” *Physical Review C* **82** (2010) 064311.
- [Vin11a] P. Vingerhoets. “Nuclear structure of Cu isotopes studied with collinear laser spectroscopy.” *Ph.D. thesis, KU Leuven, Belgium* (2011).
- [Vin11b] P. Vingerhoets, K. T. Flanagan *et al.* “Magnetic and quadrupole moments of neutron deficient $^{58-62}\text{Cu}$ isotopes.” *Physics Letters B* **703** (2011) 34–39.
- [Vos13] A. Voss, M. R. Pearson *et al.* “First Use of High-Frequency Intensity Modulation of Narrow-Linewidth Laser Light and Its Application in Determination of $^{206,205,204}\text{Fr}$ Ground-State Properties.” *Physical Review Letters* **111** (2013) 122501.
- [Vos15] A. Voss *et al.* “Nuclear moments and charge radii of neutron-deficient francium isotopes and isomers.” *Physical Review C* **91** (2015) 044307.
- [Wal89] K. Wallmeroth *et al.* “Nuclear shape transition in light gold isotopes.” *Nuclear Physics A* **493** (1989) 224–252.
- [Wan12] L. W. Wansbeek, S. Schlessner, B. K. Sahoo *et al.* “Charge radii of radium isotopes.” *Physical Review C* **86** (2012) 015503.
- [Wan17] M. Wang *et al.* “The AME2016 atomic mass evaluation (II). Tables, graphs and references.” *Chinese Physics C* **41** (2017) 030003.
- [Web05] C. Weber *et al.* “Weighing excited nuclear states with a Penning trap mass spectrometer.” *Physics Letters A* **347** (2005) 81–87.
- [Wei02] L. Weissman, J. Cederkall *et al.* “ β^+ decay of ^{61}Ga .” *Physical Review C* **65** (2002) 044321.
- [Wel17] A. Welker *et al.* “Binding Energy of ^{79}Cu : Probing the Structure of the Doubly Magic ^{78}Ni from Only One Proton Away.” *Physical Review Letters* **119** (2017) 192502.
- [Wie13] F. Wienholtz *et al.* “Masses of exotic calcium isotopes pin down nuclear forces.” *Nature* **498** (2013) 346–349.
- [Wig27] E. Wigner. “Einige Folgerungen aus der Schrödingerschen Theorie für die Termstrukturen.” *Zeitschrift für Physik* **43** (1927) 624–652.

- [Wil17a] S. G. Wilkins. “Collinear resonance ionization spectroscopy of exotic francium and radium isotopes.” *Ph.D. thesis, The University of Manchester, Manchester, United Kingdom* (2017).
- [Wil17b] S. G. Wilkins, K. M. Lynch *et al.* “Quadrupole moment of ^{203}Fr .” *Physical Review C* **96** (2017) 034317.
- [Wil19] Shane G. Wilkins. “Private communication.” (2019).
- [Woh70] F. K. Wohn, W. L. Talbert Jr. and J. K. Halbig. “The decay of ^{85m}Kr .” *Nuclear Physics A* **152** (1970) 561–569.
- [Wra17] C. Wraith, X. F. Yang, L. Xie *et al.* “Evolution of nuclear structure in neutron-rich odd-Zn isotopes and isomers.” *Physics Letters B* **771** (2017) 385–391.
- [Xie19] L. Xie, X. F. Yang, C. Wraith *et al.* “Nuclear charge radii of $^{62-80}\text{Zn}$ and their dependence on cross-shell proton excitations.” *Physics Letters B* **797** (2019) 134805.
- [Xu19] X. Xu *et al.* “Masses of neutron-rich $^{52-54}\text{Sc}$ and $^{54,56}\text{Ti}$ nuclides: The $N = 32$ subshell closure in scandium.” *Physical Review C* **99** (2019) 064303.
- [Yan16a] X. F. Yang *et al.* “Study of neutron-rich $^{52,53}\text{K}$ isotopes by the measurement of spins, moments and charge radii.” *CERN-INTC* P-458 (2016).
- [Yan16b] X. F. Yang, C. Wraith, L. Xie *et al.* “Isomer Shift and Magnetic Moment of the Long-Lived $1/2^+$ Isomer in $^{79}_{30}\text{Zn}_{49}$: Signature of Shape Coexistence near ^{78}Ni .” *Physical Review Letters* **116** (2016) 182502.
- [Yan18a] K. Yanase *et al.* “Shell-model study of Pb, Bi, Po, At, Rn, and Fr isotopes with masses from 210 to 217.” *Physical Review C* **98** (2018) 014308.
- [Yan18b] X. F. Yang, Y. Tsunoda *et al.* “Investigating the large deformation of the $5/2^+$ isomeric state in ^{73}Zn : An indicator for triaxiality.” *Physical Review C* **97** (2018) 044324.
- [Yor16] D. T. Yordanov *et al.* “Simple Nuclear Structure in $^{111-129}\text{Cd}$ from Atomic Isomer Shifts.” *Physical Review Letters* **116** (2016) 032501.
- [Zaw85] D. Zawischa. “On the origin of odd-even-staggering of nuclear charge radii.” *Physics Letters B* **155** (1985) 309–312.
- [Zha15] J. Zhang, M. Tandecki, R. Collister *et al.* “Hyperfine Anomalies in Fr: Boundaries of the Spherical Single Particle Model.” *Physical Review Letters* **115** (2015) 042501.

FACULTY OF SCIENCE
DEPARTMENT OF PHYSICS AND ASTRONOMY
INTERDISCIPLINARY RESEARCH

Celestijnenlaan 200D box 2418

B-3001 Leuven

gregoryjames.farooqsmith@kuleuven.be

<https://www.fys.kuleuven.be/iks/interdisciplinary-research>

