

Laser-assisted decay and optical spectroscopy studies of neutron-deficient thallium isotopes.

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Preface

This thesis describes decay- and laser-spectroscopy studies of neutron-deficient $^{179-184}\text{Tl}$ isotopes that were performed at the ISOLDE facility. Chapter 1 provides an introduction to the nuclear shell and liquid drop model and to nuclear deformation and motivates why the neutron-deficient thallium isotopes are interesting to study. The principles and experimental observables of α and γ decay and laser spectroscopy are introduced in Chapter 2 and in Chapter 3, experimental details related to the setup are given. Chapter 4 is the main chapter of this thesis and provides two papers in which the decay spectroscopy results obtained in this work are published:

C. Van Beveren, A. N. Andreyev, A. E. Barzakh, T. E. Cocolios, D. Fedorov, V. N. Fedosseev, R. Ferrer, M. Huyse, U. Köster, J. Lane, V. Liberati, K. M. Lynch, B. A. Marsh, T. J. Procter, E. Rapisarda, K. Sandhu, M. D. Seliverstov, P. Van Duppen, M. Venhart, M. Veselský.

Internal decay characteristics of ^{184}Tl

Physical Review C **92**, 014325 (2015)

C. Van Beveren, A. N. Andreyev, A. E. Barzakh, T. E. Cocolios, R.P. de Groote, D. Fedorov, V. N. Fedosseev, R. Ferrer, L. Ghys, M. Huyse, U. Köster, J. Lane, V. Liberati, K. M. Lynch, B. A. Marsh, P. L. Molkanov, T. J. Procter, E. Rapisarda, K. Sandhu, M. D. Seliverstov, P. Van Duppen, M. Venhart, M. Veselský.

α -decay study of $^{182,184}\text{Tl}$

Journal of Physics G: Nuclear and Particle Physics **43**, 025102 (2016)

Next to these papers, Chapter 4 contains a more detailed description of a dead time problem encountered when analysing the data. In Chapter 5 the results of the laser-spectroscopy study are presented and discussed. Finally, Chapter 6 concludes on the results that were achieved in this thesis and provides an outlook into the future.

Summary

The neutron-deficient thallium isotopes with one proton less than the $Z = 82$ shell closure, are situated in an interesting region of the nuclear chart, notorious for intruder states and shape coexistence. Shape coexistence is the remarkable phenomenon in which two or more distinct types of deformation occur at low energy in the same atomic nucleus. Shape coexistence has been studied intensively, experimentally as well as theoretically in different nuclei in the light-lead region and the isomerism in the thallium isotopes was among the first indications of this phenomenon. Different shapes, whose structure has been linked to specific proton orbitals above and below the $Z = 82$ shell closure, are present at low energy in the neutron-deficient odd-mass thallium nuclei. In the odd-odd nuclei, the coupling of an unpaired proton and unpaired neutron gives rise to multiplets of low-lying states from which some can be isomeric. Since thallium has one proton missing in the major proton shell, and when approaching neutron mid-shell at $N = 104$, the structure of these isotopes manifests itself by the interplay between shape coexistence phenomena as well as single-particle properties.

In this thesis, the structure of a number of neutron-deficient thallium isotopes is studied with laser-assisted decay and optical spectroscopy performed at the ISOLDE facility in CERN (Geneva, Switzerland). This dual approach was essential to identify the different decay branches and disentangle the decay schemes of the different long-lived states in the thallium isotopes. The decay products were detected with the Windmill system and two HPGe detectors surrounding the Windmill chamber.

A study of the internal decay of the (10^-) intruder state in ^{184}Tl revealed an excitation energy of 506.1(1) keV, extending the $(10^-) \rightarrow (7^+)$ energy systematics beyond neutron mid-shell and confirming the parabolic behavior of the excitation energy as a function of neutron number. Using the extracted half-life of the (10^-) state, the retardation factors of the depopulating E3 and M2 transitions were determined and compared with retardation factors in neighboring odd-mass and

even-mass thallium isotopes. Combined with the information on the published reduced α -decay widths of the bismuth isotopes populating levels in neighboring thallium nuclei, a confirmation of the proton 1p-2h intruder character of the (10^-) isomer and an interpretation of the levels in ^{184}Tl fed in the isomeric decay in terms of $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ and $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configurations could be obtained. From the results of the laser-spectroscopy study, it was observed that the magnetic moment of the (7_1^+) state shows increased importance of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration, compared to respective magnetic moments in the heavier odd-odd thallium isotopes. The similarity of the charge radius of the (10^-) isomeric state with the charge radii of the $\pi h_{9/2}$ -based intruder states in heavier odd and odd-odd thallium isotopes supports the proton intruder character of this state in ^{184}Tl .

Through the observation of hindered and unhindered α decay, low-lying states, possibly intruders, in the daughter nuclei can be selectively studied and identified, yielding direct information on the excitation energy, decay pattern and possible configurations involved. In this thesis, the results of an α decay of $^{182,184}\text{Tl}$ are presented. New fine-structure α decays have been observed for both isotopes and previous observed α -decay lines were confirmed. Using a purification procedure on the singles α -decay energy spectra, α -decay branching ratios could be determined for the three long-lived (10^-) , (7^+) and (2^-) states in ^{184}Tl . Also for ^{182}Tl new α -decay lines have been identified and a lower limit of the α -decay branching ratio could be determined. Using α - γ coincidence analysis, multiple γ rays were observed de-exciting levels in $^{178,180}\text{Au}$ fed by $^{182,184}\text{Tl}$ α decays. The γ transitions connecting these low-lying states in $^{178,180}\text{Au}$ are essential to sort the data and possibly identify bands from in-beam studies in these isotopes. Owing to the complex fine-structure α decays and limited knowledge about the structure of the daughter nuclei, only partial level schemes could be constructed for both gold isotopes in the present work. Reduced α -decay widths have been calculated, using the determined α -decay branching ratios and half-lives of the different isomeric states, and are compared with values obtained in neighboring odd- and even-mass thallium isotopes. Except for the allowed α decay of the ^{184}Tl (10^-) state, the other fine-structure α decays observed in this study are hindered. This points to strong structural changes between parent thallium and daughter gold isotopes.

In laser-spectroscopy studies, the nuclear states themselves are probed, providing model-independent information on ground and isomeric states through the measurement of the hyperfine structure patterns in long chains of isotopes. In this work, $^{179-184}\text{Tl}$ has been studied using the in-source laser spectroscopy technique with the aim to extend the previously known isomeric chain beyond neutron mid-shell at $N = 104$. The deduced magnetic moments point to a smooth isotopic change in nuclear structure at neutron mid-shell and beyond.

A similar smooth behavior has been observed for the extracted charge radii testifying to the persistence of the near-spherical shape of the ground state, comparable to the previously observed trend in the lead isotopes. The newly measured charge radii of the thallium isomeric states involving the $\pi h_{9/2}$ intruder orbital are significantly larger than those of the ground states.

Beknopte samenvatting

De neutron-arme thallium isotopen, met één proton minder dan de $Z = 82$ schilsluiting, bevinden zich in een interessant gebied van de kernkaart dat gekend is voor vormcoëxistentie en zogenaamde indringstructuren. Vormcoëxistentie is het opmerkelijke fenomeen waarbij twee of meer toestanden met vergelijkbare excitatie-energie voorkomen met een verschillend intrinsiek vervormingstype. Dit fenomeen is grondig bestudeerd in verscheidene kernen in het lichte-lood gebied, zowel op experimenteel als theoretisch vlak en het isomerisme in de thallium kernen was één van de eerste indicaties voor vormcoëxistentie. In de neutron-arme oneven thallium kernen bleken verscheidene vormen aanwezig te zijn bij lage excitatie-energie en deze vormen werden gelinkt aan specifieke proton orbitalen van onder en boven de $Z = 82$ schilsluiting. In de oneven-oneven kernen ontstaan er multipletten van laag-gelegen toestanden, in sommige gevallen isomeren, door de koppeling tussen het ongepaarde proton en het ongepaarde neutron. Vermits thallium één proton mist in de hoofdschil, zal de structuur van deze kernen bepaald worden door een wisselwerking van vormcoëxistentie fenomenen en één-deeltjes eigenschappen.

In deze thesis werd de structuur van een aantal neutron-arme thallium kernen bestudeerd door gebruik te maken van laser-geassisteerde verval en optische spectroscopie experimenten uitgevoerd aan de ISOLDE faciliteit in CERN (Genève, Zwitserland). Deze tweevoudige benadering was essentieel om de verscheidene verval takken te identificeren en om de verval schema's van de verschillende lang-levende toestanden te ontrafelen. De vervalproducten werden gedetecteerd met het Windmill systeem met errond twee HPGe detectoren.

Uit een studie van het intern verval van de (10^-) indringtoestand in ^{184}Tl werd een excitatie-energie van 506.1(1) keV bepaald voor deze toestand. Met dit resultaat werd de energie-systematiek voor de $(10^-) \rightarrow (7^+)$ overgangen uitgebreid voorbij het midden van de neutron schil en wordt het parabolisch gedrag van deze excitatie-energie in functie van het aantal neutronen bevestigd. Met de bepaalde halfwaardetijd voor de (10^-) toestand konden

de vertragsfactoren van de E3 en M2 overgangen, die vertrekken van deze toestand, bepaald worden. De op die manier bepaalde vertragsfactoren werden vergeleken met deze in naburige oneven en even thallium isotopen. De resultaten bekomen uit de studie van het intern verval werden ook gecombineerd met informatie over de reeds gepubliceerde gereduceerde alfa-verval breedtes van naburige bismut isotopen. In het α verval van deze bismut isotopen worden gelijkaardige toestanden in naburige thallium kernen gevoed, waardoor het proton één deeltje-twee gaten indringer karakter van de (10^-) isomeer in ^{184}Tl kon bevestigd worden. De toestanden die in het intern verval van de (10^-) toestand gevoed worden, werden ook geïnterpreteerd aan de hand van de $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ and $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuraties. De resultaten van de laser spectroscopische studie wijzen op een toenemende bijdrage van de $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuratie in het magnetisch moment van de (7_1^+) toestand, relatief ten opzichte van de magnetische momenten van de zwaardere oneven-oneven thallium isotopen. Het gelijkaardige gedrag van de ladingsstraal van de (10^-) isomere toestand met deze van de $\pi h_{9/2}$ -gebaseerde indringtoestanden in de zwaardere oneven en oneven-oneven thallium isotopes ondersteunt het indring karakter van deze toestand in ^{184}Tl .

Door de observatie van gehinderd en ongehinderd α verval, kunnen laag-gelegen toestanden, mogelijk indringtoestanden, in de dochter kernen geïdentificeerd en bestudeerd worden. Hierdoor wordt rechtstreeks informatie bekomen over de excitatie-energie, het verval pad en de mogelijke configuraties die betrokken zijn bij het verval proces. In deze thesis worden de resultaten van een α -verval studie van $^{182,184}\text{Tl}$ voorgesteld. Voor beide isotopen werden tot nu toe onbekende α -verval lijnen geïdentificeerd. Door een zuiveringsprocedure op de spectra konden de individuele α -verval takken van de drie lang-levende (10^-) , (7^+) en (2^-) toestanden in ^{184}Tl bepaald worden. Voor ^{182}Tl werd een onderlimiet bepaald voor de α -verval vertakkingsverhouding. Aan de hand van α - γ coincidentie analyse konden meerdere γ overgangen geobserveerd werden die toestanden in $^{178,180}\text{Au}$ de-exciteren en gevoed worden door het α verval van $^{182,184}\text{Tl}$. Deze γ overgangen, die meer inzicht geven in de laag-gelegen toestanden in $^{178,180}\text{Au}$, zijn essentieel om de data te sorteren alsook om mogelijke banden te identificeren in *in-beam* studies van deze isotopen. In dit werk konden enkel partiële vervalschema's bepaald worden voor beide isotopen. De reden hiervoor is dat er slechts weinig bekend is over de structuren van de dochter kernen, alsook door het complexe karakter van het α verval van de oneven-oneven thallium isotopen. Met de bepaalde α -verval vertakkingsverhoudingen en de halfwaardetijden van de verschillende isomere toestanden, konden de gereduceerde α -verval breedtes berekend worden. Deze resultaten werden vergeleken met de waardes bekomen in naburige even en oneven thallium isotopen. Enkel het α verval van de (10^-) toestand blijkt ongehinderd te zijn, de andere α -verval takken geobserveerd in deze studie zijn allemaal gehinderd.

Dit duidt op sterke structuur veranderingen tussen de thallium moeder en goud dochter isotopen.

Bij een laser-spectroscopie studie gaat men de toestanden in een kern aftasten door de hyperfijn structuur op te meten in lange reeksen van isotopen. Hierdoor wordt kernmodel-onafhankelijke informatie bekomen over de grond en isomere toestanden. In dit werk werden $^{179-184}\text{Tl}$ isotopen bestudeerd met laser spectroscopie. Het doel van deze studie was de reeds bekende isomere reeks uit te breiden naar de meer neutron-arme isotopen, voorbij het midden van de neutron schil. Uit de bepaalde magnetische momenten kon vastgesteld worden dat de structuur van de isotopen gelijkmatig veranderd voorbij het middel van de neutron schil. Eenzelfde trend werd afgeleid voor de ladingsstralen die duidelijk vasthouden aan het bijna-sferisch gedrag geobserveerd in de zwaardere isotopen. Het gedrag van de thallium ladingsstralen vertoont een gelijkaardig gedrag als de lood isotopen. De in dit werk opgemeten ladingsstralen van de isomere toestanden in thallium gebaseerd op de $\pi h_{9/2}$ indring orbitaal, zijn duidelijk groter dan deze van de grond toestand en volgen hierdoor de systematiek eerder opgemeten in de zwaardere isotopen.

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1 | Introduction

In 1911 Rutherford, who had demonstrated with his α -particle scattering experiments that the mass of an atom is mainly concentrated in a very small positively charged nucleus, launched the beginning of a new branch in physics research, i.e. nuclear physics. It was discovered that the atomic nucleus consists of A nucleons: Z protons and N neutrons. These nucleons interact with each other in a complicated way and can bind in nuclei in various combinations. As a consequence, nuclei possess a rich and fascinating structural diversity, which provides a powerful probe of the fundamental forces in nature. The interaction keeping the nucleons together is the strong interaction, which can overcome the electromagnetic repulsion between the positively charged protons. Up to present no exact description of this fundamental force exists.

In order to get a better understanding of the forces that govern the evolution and behavior of matter, the structure of nuclei is studied with many different experimental techniques, yielding experimental data that can be used to validate theories and give guidance to further developments. A widely used technique is the study of radioactivity of unstable isotopes. In these studies information on the properties of the nuclear states of mother and daughter can be obtained.

Due to the difficulties describing many particle systems like a nucleus from first principles, one has to rely on approximations using nuclear models for the descriptions. Throughout the years different theoretical descriptions of the nucleus, that simplify the complex many-body problem in their own way, have been developed. The nuclear shell model, where the nucleons are considered as particles moving independently of each other in a central nuclear potential, interacting via the residual interaction and where specific numbers of protons and neutrons give rise to exceptionally stable nuclei, is still very successful. This stability is explained by the fact that a particular amount of protons or neutrons leads to the filling of a “shell” of energy levels. Near stability the so-called “magic” numbers are situated at $N, Z = 2, 8, 20, 28, 50, 82, 126$ (only known for N), $\dots$ The shell model is suited for the description of nuclei with few valence

nucleons outside the closed shells, since the basic assumption in the shell model is that the nucleus can be described in first approximation as an ensemble of particles moving independently in a spherical mean field, interacting via the residual interaction. In nuclei with many valence nucleons of both types, the interaction between protons and neutrons gradually builds up. The nucleus tends to deviate from its spherical shape and end up in an energetically more favorable deformed shape. Collective phenomena such as rotation and vibration, involving many nucleons, emerge.

1.1 Shape coexistence

For certain numbers of protons and neutrons the phenomenon called shape coexistence [1, 2, 3], where two or more distinct types of deformation occur at low energy in the same atomic nucleus, is manifested in the nuclear landscape. Shape coexistence is governed by the interplay between two opposing tendencies. On one side the stabilizing effect of closed shells and subshells, which causes the nucleus to retain a spherical shape and leads to a level structure based on a redistribution of the protons and/or neutrons over the spherical shell orbitals. On the other side, the residual interactions between protons and neutrons, or, the correlation energy gain, in which the proton-neutron interaction energy is a major contribution. This interaction is mainly multiplicative in the number of interacting protons times the number of interacting neutrons and drives the nucleus into a deformed shape. Coexistence in nuclei has been a feature of nuclear structure for over 50 years and appears to occur in all nuclei. Understanding the occurrence of shape coexistence in atomic nuclei is an important challenge faced by theories of nuclear structure. The subject has advanced a lot over the years due to an important reciprocity between experimental observation and theoretical description. For an overview on the experimental evidence and the recent progress in theoretical descriptions, we refer to [4].

In a number of nuclei, it becomes possible to deform the nucleus and lower its total energy considerably, relative to the spherical ground state configuration, when excitation of nucleons across a major shell occurs, giving rise to so-called intruder states. These deformed intruder states then coexist with the spherical ground state. In the deformed Nilsson model (see chapter 2) the strong up- and downslowing orbitals just above and below the closed shells show already in an intuitive way the natural tendency for nuclei to acquire a deformed shape next to the spherical shape corresponding to the closed shell itself.

Shape coexistence in nuclei can be identified by a number of specific experimental fingerprints, such as retarded electromagnetic transitions (see chapters 2 and 4).

For an overview of the spectroscopic fingerprints of shape coexistence, we refer e.g. to [1] and [5].

1.2 The neutron-deficient thallium isotopes

1.2.1 Level structure

The region of the neutron-deficient isotopes at and near the $Z = 82$ proton shell closure has drawn considerable interest as it exhibits a clear manifestation of shape coexistence. The study of this region has been challenging since it is centered on isotopes that lie far from β stability. The isomerism in the thallium isotopes was among the first indications of shape coexistence in this region of the nuclear chart. Different shapes, whose structure has been linked to specific proton orbitals above and below the $Z = 82$ shell closure, are present at low energy in the neutron-deficient odd-mass thallium nuclei. Next to spherical proton-hole configurations, involving the “regular” $\pi 3s_{1/2}$ and $\pi 2d_{3/2}$ orbitals, one particle-two hole (1p-2h) configurations, involving the $\pi 1h_{9/2}$ and $\pi 1i_{13/2}$ orbitals from above the $Z = 82$ shell closure are identified. Rotational band structures built on top of the latter, so-called intruder states, were observed (see references 1 to 7 in [6] and Fig. 18 in [4]). In Fig. 1.1 an overview is given of the shell model orbitals around $Z = 82$ for protons (π) and around $N = 104$ for neutrons (ν). The energy of the $\pi 1p\text{-}2h$ band heads shows a typical parabolic behavior as a function of neutron number, reaching a minimum at $N = 108$, close to $N = 104$, mid-shell between $N = 82$ and $N = 126$ (see Fig. 1.2). This leads for the case of the $(9/2^-)$ state to isomerism. For $^{185,191,193,195,197}\text{Tl}$, the mean-square charge radii of the $(1/2^+)$ ground state as well as the $(9/2^-)$ isomer were measured. A large isomer shift was observed, implying a larger deformation of the $(9/2^-)$ isomer relative to that of the $(1/2^+)$ ground state [7].

The occurrence of intruder states and shape coexistence in odd-mass thallium isotopes, raises the question of where such states appear in the even-mass thallium isotopes and what role the unpaired neutron plays when coupled to the unpaired intruder proton. In [7], the isomer shift of the intruder-based $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ state, relative to the regular $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ ground state, has been determined in ^{186}Tl . This is the first time the intruder isomer shift has been measured in the odd-odd thallium isotopes (see also section 1.2.2). Bands based on isomeric states have been observed also in doubly-odd thallium isotopes in the mass range $A = 186\text{--}200$ [6]. These bands represent examples of collective structures based on high-spin excitations in odd-odd nuclei and the band heads were successfully described as semidecoupled $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ multiplets by using a two-quasiparticle plus rotor model [6]. Isomeric transitions

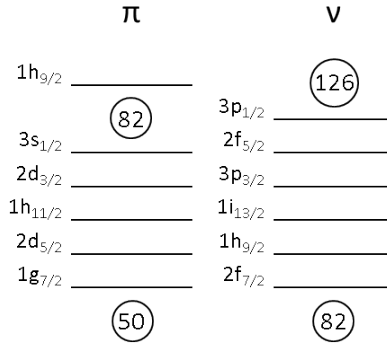


Figure 1.1: Overview of the shell model orbitals around $Z = 82$ for protons (π) and around $N = 104$ for neutrons (ν).

of E3, M2, E1/M2 and E1 character, depopulating the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ band heads towards the regular $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ states, were found using in-beam and decay spectroscopy. The changes in multipolarity reflect successive changes in the band head spin values corresponding to level crossings within the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ multiplet. These level crossings are illustrated in Fig. 1.3.

In [10] Paar derived an expression for the energy splitting within a given proton-neutron multiplet resulting in a quadratic dependence on the spin $I(I+1)$. Using more advanced calculations within the framework of the proton-particle neutron-quasiparticle quadrupole vibrator model, the authors of [11] looked into possible configurations and energy splittings in the odd-odd thallium and bismuth nuclei. Using an explicit proton-neutron interaction, approximated by a zero-range force including spin exchange in addition to the proton-neutron orbitals coupling to low-lying quadrupole vibrations of the underlying core, the authors of [11] succeeded in retrieving the well-known parabolic rule from Paar. The authors also showed that for $A(\text{Tl}) < 188$, the 10^- state from the intruder based $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ multiplet will stay below the 8^- , 9^- and 11^- states and thus possibly become isomeric. This is in contrast to the situation for $A(\text{Tl}) > 188$ where the 8^- is expected to be the lowest state (see Fig. 8 in [11]). These level crossings were also predicted in [12] using the two-quasiparticle plus rotor model [6]. The authors of [12] concluded that the behavior of the $8^- \rightarrow 10^-$ multiplet of states appeared to be established mainly by the binding to the quadrupole field of the core and that the proton-neutron force did not seem to play a considerable role in determining their structure. However, in [11] the authors were able to differentiate more precisely between effects of the quadrupole phonon coupling and the residual valence proton-neutron interaction.

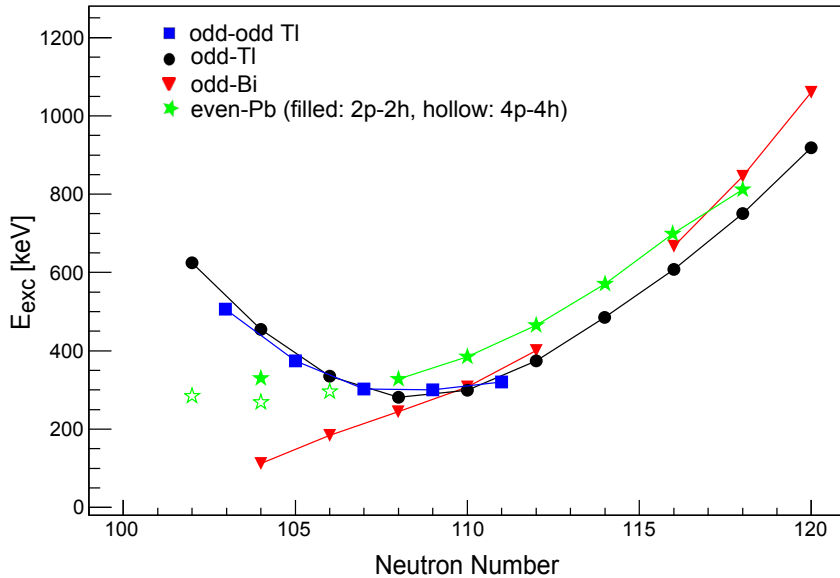


Figure 1.2: [Color] Systematics of the odd-Tl ($9/2^- \rightarrow 1/2^+$) and odd-odd Tl ($10^- \rightarrow 7^+$) $\pi(1p-2h)$, odd-Bi ($1/2^+ \rightarrow 9/2^-$) $\pi(2p-1h)$ and even-Pb ($0_2^+ \rightarrow 0_1^+$ or $0_3^+ \rightarrow 0_1^+$) (filled: $\pi(2p-2h)$, hollow: $\pi(4p-4h)$) intruder-state excitation energies as a function of neutron number (the even-Pb excitation energies are divided by two). The strong correlated behavior is further discussed in the text (see section 1.2.3). Data are from [8, 9] and this work. It should be noted that these are the assumed $\pi p-h$ configurations.

The neutron-deficient region around the $Z = 82$ proton shell closure exhibits many α -decaying isotopes and the observation of hindered and unhindered α decay has proven to be a powerful method to selectively study and identify intruder states in nuclei [14, 15, 16]. From β -decay and in-beam studies a wealth of information can be obtained such as level energies, feeding and decay patterns, spin and parities and transition probabilities. All this experimental information however does not give direct access to the underlying structure and theoretical guidance is needed to interpret the data. Often, α -decay spectroscopy avoids the complexity encountered when using in-beam or β -decay studies because the α -decay energy differences impose stringent constraints on the assignment of internal transitions via coincidence spectroscopy. Alpha decay can be used to probe specific structures and determine the energy and transition probabilities, but is also sensitive to the specific shell-model orbitals

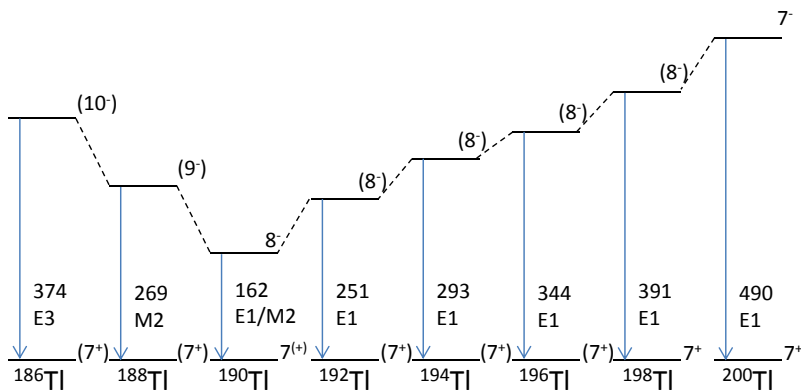


Figure 1.3: Systematics of isomeric transitions depopulating the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ band heads in odd-odd $^{186-200}\text{Tl}$. The observed energy (in keV) and multipolarity of each transition are indicated. Data are from [12, 13, 14].

that are involved in the decay. Low-lying states are strongly populated due to the strong Q-value dependence of the quantum-mechanical tunneling probability through the Coulomb barrier. Therefore in even-even nuclei, low-lying 0^+ band heads, that are often not observed in in-beam studies due to out-of-band decay or are missed in β -decay studies due to the weak feeding, can be populated and studied in α decay. Furthermore, the formation probability of the α particle, expressed by the reduced α -decay width, is extremely sensitive to the overlap in wave functions between the initial state and final state plus α particle, strongly enhancing $\Delta\ell = 0$ transitions between states of similar origin. Alpha-decay studies have led to the characterization of intruder states in the $\text{Rn} \rightarrow \text{Po} \rightarrow \text{Pb} \rightarrow \text{Hg} \rightarrow \text{Pt}$ and $\text{At} \rightarrow \text{Bi} \rightarrow \text{Tl}$ chains ([4, 16] and references therein).

1.2.2 Spins, moments and radii

Atomic spectroscopy uniquely provides additional and, in a number of cases, model-independent information on ground and isomeric state properties through the hyperfine interaction between the electrons and the nucleus. The first indication of shape coexistence in the region around $Z = 82$ came from the observation of a large isomer shift for $^{185,187}\text{Tl}$ in optical hyperfine structure studies of the mercury ($Z = 80$) isotopes [17]. This was interpreted as a dramatic change in shape from a weakly-deformed oblate shape in the heavier isotopes to

a more-strongly deformed prolate shape [18]. Since then, many isotopic chains of mean-square charge radii have been measured. An overview of the mean-square charge radii in the lead-region available from literature is given in Fig. 1.4. Both odd- and even-neutron gold isotopes ($Z = 79$) change their shapes from weakly oblate to strongly prolate deformed after $N = 107$ due to the influence of the $\pi h_{9/2}$ intruder orbital [19]. In the case of the lead isotopic chain, the isotope shifts indicate that the ground state remains spherical down to and beyond neutron mid-shell at $N = 104$ [20, 21]. The polonium and platinum isotopes depart from this spherical trend for $N < 116$, although the magnitude of the deformation in the platinum isotopes is not as important as in the polonium case. The thallium isotopes reveal the presence of near-spherical ground states as well as both oblate and prolate configurations (see e.g. [22, 23]). For $^{185,191,193,195,197}\text{Tl}$, the mean-square charge radii of the $(1/2^+)$ ground state as well as the $(9/2^-)$ isomer were measured. A large isomer shift was observed, implying a larger deformation of the $(9/2^-)$ isomer relative to that of the $(1/2^+)$ ground state [7]. A similar isomer shift was also found for the odd-odd isotope ^{186}Tl . The literature data for the magnetic dipole moments of the thallium isotopes are given in Fig. 3 of Ref. [7]. The magnetic moments of the different thallium nuclear states follow smooth isotopic trends, which points to the absence of abrupt structural changes in these nuclei. An overview of the currently measured electric quadrupole moments for the thallium isotopes with $N > 115$ can be found in [24]. Spin assignments in the lighter thallium isotopes ($N < 109$) are based on systematics of the heavier isotopes. These tentative assignments are strongly supported by the results of different decay and laser spectroscopy studies, e.g. favored/unfavored α decay in bismuth mother isotopes and reasonable/unreasonable magnetic dipole moments in view of systematics.

1.2.3 Theoretical considerations

A remarkable parabolic behavior of the intruder excitation energy as a function of neutron number has been observed in the nuclei in the lead region through the intrusion of configurations across the $Z = 82$ closed shell. This parabolic pattern with a minimum near neutron mid-shell, shown in Fig. 1.2, illustrates that collectivity is largely related to the number of valence nucleons outside closed shells. From a shell-model description of intruder states using the mechanism of particle-hole excitations across closed shells, evidence for a scaling of the intruder excitation energy with the number of particles and hole pairs formed has been deduced [25]. Instead of calculating the intruder energy from a given nuclear hamiltonian, using effective or more realistic residual interactions, the authors have separated the major components of the nucleon-nucleon interaction

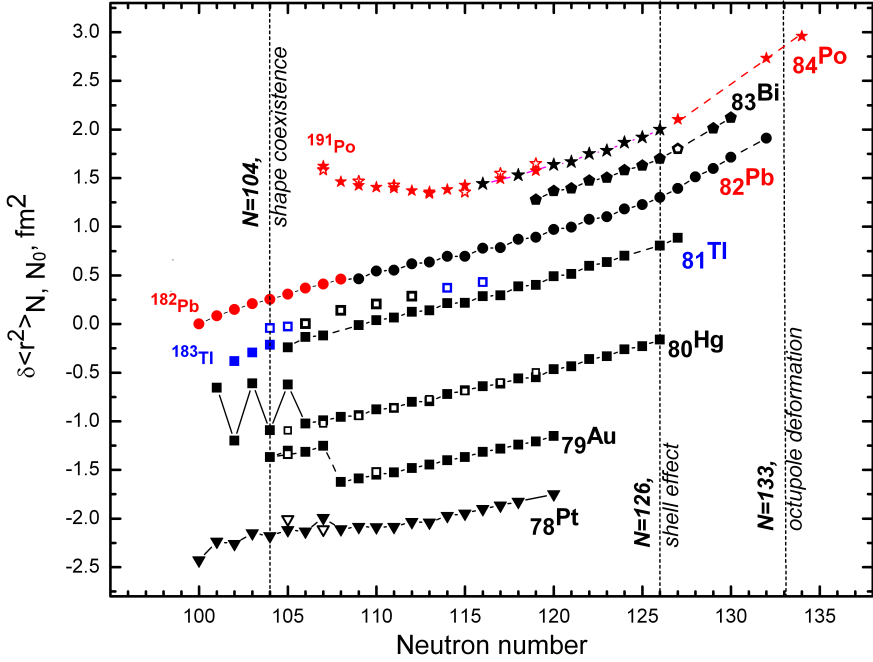


Figure 1.4: [Color] Systematics in mean-square charge radii $\delta \langle r^2 \rangle$ for the lead region as a function of neutron number. Full and hollow symbols are for ground and isomeric states respectively. The blue symbols represent data measured at the IRIS facility, the red symbols at the ISOLDE facility. Black symbols are older literature data collected at different facilities. Data are from [8].

and their corresponding contribution to the intruder state excitation energy. They focused on the pairing correlations amongst identical nucleons and the long-range attractive component between protons and neutrons, more specific the quadrupole interaction. Although major simplifications in the detailed form of the residual nucleon-nucleon interaction have been introduced, general features for the nuclear structure at low energy remain. The resulting scaling law is given by:

$$\frac{1}{2}E_{intr}(0^+; even) \simeq E_{intr}(j_\pi; odd) \simeq E_{intr}(j_\pi j_\nu; I, odd - odd) \quad (1.1)$$

The above scaling law is valid for the particular member of the intruder multiplet $(j_\pi j_\nu; I, odd - odd)$ for which the two-body proton-neutron matrix element is almost equal to the ground state multiplet matrix element $(j_{\pi'} j_{\nu'}; I_{gs}, odd - odd)$:

$$\langle (j_\pi j_\nu) I | V_{\pi\nu} | (j_\pi j_\nu) I \rangle - \langle (j_{\pi'} j_{\nu'}) I_{gs} | V_{\pi\nu} | (j_{\pi'} j_{\nu'}) I_{gs} \rangle \simeq 0 \quad (1.2)$$

The scaling law has been extensively studied in [13, 14, 25] and for more details we refer the reader to these papers.

In Fig. 1.2 the energy systematics for the lowest intruding coexisting structures in the odd-Tl and odd-odd Tl, odd-Bi and even-Pb isotopes are shown. For the odd-odd Tl isotopes, the energy difference of the (10^-) $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}] \rightarrow (7^+)$ $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ is used, since the difference of the matrix elements for the spin (10^-) member of the multiplet with the matrix elements of the spin (7^+) ground state is almost zero. This can be seen in Fig. 5 of Ref. [25]. The excitation energies for the even-Pb isotopes in Fig. 1.2 are divided by two. One can see that the thallium $\pi(1p-2h)$ and bismuth $\pi(2p-1h)$ excitation energies are almost exactly one-half of the lead $\pi(2p-2h)$ excitation energies. This is consistent with the energies scaling as the number of correlated proton-pairs and nicely illustrates equation (1.1). The candidate $\pi(4p-4h)$ states are shown as hollow green stars. From $N < 108$ the parabolic behavior breaks down for the bismuth isotopes. The minimum of the parabolic pattern is situated exactly at neutron mid-shell $N = 104$ for the even-Pb isotopes. This is not the case for the odd- and even-Tl isotopes or even-Hg isotopes (see e.g. Fig. 1 in Ref. [26]) where the minima are situated at $N = 108$, $N = 109$ and $N = 102$ respectively. The reason why these minima for certain isotopes are exactly at $N = 104$ and for others not, is not understood up to present.

A complementary theoretical description for intruder states and shape coexistence is given by the mean-field approach. This approach results in energy surfaces with (sometimes multiple) minima that correspond to deformed density distributions in many cases. The model can also include the strong pairing correlations that are active in the nucleus, simultaneously optimizing both the mean single-particle field and the pairing (mean-pair field) properties in nuclei. As a result, coexisting shapes may appear for certain proton and neutron numbers. A well-known example of such a energy surface with different coexisting minima is illustrated in Fig. 1.5 for ^{186}Pb [27].

Shape coexistence in the neutron-deficient lead region cannot be described on the level of mean-field models in a fully satisfactory way. To obtain spectroscopic information, one has to go beyond a strict mean-field approach. Low-lying excited states are mixed into the calculated mean-field ground state which can be removed by configuration mixing, i.e., superposition of several mean-field states. The authors of [28] studied shape coexistence in ^{186}Pb using this method by incorporating configuration mixing of the angular momentum and particle-number projected mean-field states. For an extended review on (beyond)-mean-field descriptions for nuclear structure, we refer the reader to [29].

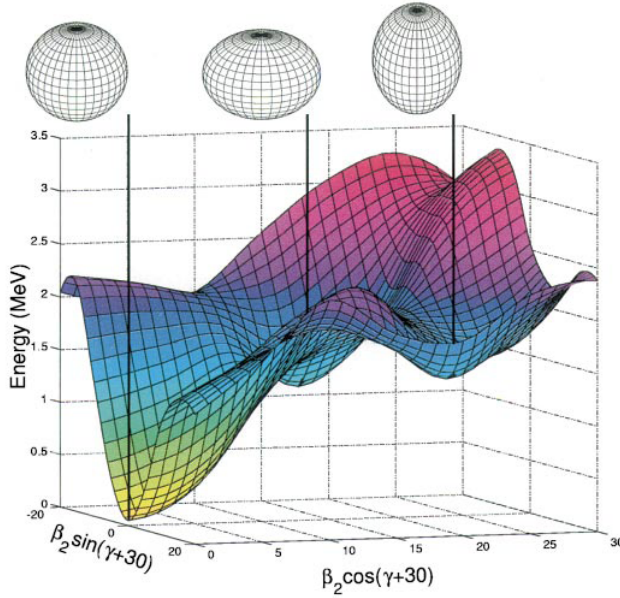


Figure 1.5: [Color] Calculated potential energy surface of ^{186}Pb . Spherical, oblate and prolate minima are indicated by thick vertical black lines. Figure is from [27].

1.2.4 Motivation and methodology

The objectives of this work are to extend the spectroscopic knowledge of the thallium isotopes towards the interesting mid-shell region by studying their decay properties and by performing laser spectroscopy measurements. Combining the high selectivity of the RILIS [30, 31, 32] with decay spectroscopy and the in-source laser spectroscopy method, exotic thallium isotopes down to $N = 97$ have been studied at ISOLDE [33] using the Windmill detection system [34, 35, 36]. Complementary decay data on isomerically purified sources were collected where this dual approach was essential to identify the different decay branches and disentangle the decay schemes of the different long-lived states in the thallium isotopes. In this work, we will focus on the internal decay characteristics of the (10^-) intruder state in ^{184}Tl , the α -decay study of $^{182,184}\text{Tl}$ and the results of an optical spectroscopy study of $^{179-184}\text{Tl}$. The rates of the electromagnetic transitions and the rates of the different α decays in these isotopes are combined with experimental results of the mean-square charge radii, magnetic moments and spins in order to characterize the intruder or regular character of the different states.

2 | Nuclear models and spectroscopy

This chapter briefly introduces the liquid drop model, the nuclear shell model and nuclear deformation, the Nilsson model and the mean-field approach, followed by the basic properties and experimental observables of α - and γ -decay spectroscopy and laser spectroscopy.

The theory presented is largely based on the textbooks by K. Heyde [37], K.S. Krane [38], S.G. Nilsson and I. Ragnarsson [39], P. Ring and P. Schuck [40] and on the topical review by B. Cheal and K.T. Flanagan [41], unless otherwise stated.

2.1 Liquid drop model

A macroscopic approach used to describe nuclear matter is that of the liquid drop model [42]. In this model, the nucleus is assumed to be an incompressible charged liquid drop. Depending on the character of the nucleon, proton or neutron, and whether the nucleon is in the inner part of the nucleus or at the outer part, the binding is different. These effects are expressed by different terms in the semi-empirical liquid drop formula.

In this formula, the binding energy of the nucleus is parameterized as followed (in a slightly generalized form of the original von Weizsäcker formula [42]):

$$B.E. = a_v A - a_s A^{2/3} - a_C \frac{Z(Z-1)}{A^{1/3}} - a_A \frac{(A-2Z)^2}{A} + \delta(A, Z) \quad (2.1)$$

where the first two terms are the volume and surface energy, respectively [38]. The volume term is proportional to the mass number since each nucleon may interact only with its nearest neighbors in the nucleus due to the short range of the strong force. The surface term corrects the previous term to take into account that the nucleons on the surface have fewer neighbors to interact with. The third term, the Coulomb term, accounts for the repulsion between protons. The fourth, so-called asymmetry term favors nuclei with an equal number of protons and neutrons, $N = Z$. The asymmetry and Coulomb terms define the path and the width of the stability line through the $N - Z$ plane. The observed even-odd staggering effect in nuclear masses leads to the addition of a pairing energy term $\delta(A,Z) = (-\Delta, 0, \Delta)$, with $\Delta \approx 12/\sqrt{A}$, for odd-odd, odd-mass, and even-even nuclei [43].

This model very successfully described some properties of the nucleus, such as the binding energy. It fails, however, to account for the finiteness of the nucleus. This effect was introduced in the liquid droplet model of the nucleus [44, 45]. In this model the finite thickness of the nuclear surface and the finiteness of the nuclear compressibility are introduced. Furthermore, the density is allowed to smoothly decrease to zero through a diffuse surface rather than with a sharp cutoff. In the liquid drop model, the energy is a function only of the shape; the liquid droplet model expands the volume, surface and Coulomb energies, which are a function of the shape and of the proton and neutron density distributions, in Taylor series around the liquid drop model values.

One of the basic properties of a nucleus is the volume or alternatively the radius. One can distinguish between the matter radius, determined by all nucleons, and the charge radius, composed by only the protons of the nucleus. In the liquid drop model, the proton distribution is considered to be homogeneous over the nuclear volume. Thus, one assumes that the radius of the proton distribution corresponds to the radius of the mass distribution, given as $R = R_0 A^{1/3}$, where $R_0 \approx 1.2$ fm and A is the mass number. The mean square charge radius of the nucleus $\langle r^2 \rangle$ in the frame of the liquid drop model is defined as [46]:

$$\langle r^2 \rangle = \frac{3}{5} R_0^2 A^{2/3} \quad (2.2)$$

2.2 The shell model

The wave functions of the nuclear states of a nucleus can be obtained by solving the many-body Schrödinger equation

$$H\Psi = E\Psi \quad (2.3)$$

where H is the hamiltonian, Ψ the many-body wavefunction and E the total energy.

The basic assumption in the independent particle model is that each nucleon is moving in an independent way in an average field. This is only an approximation since the nucleus constitutes an A -body problem interacting via the nucleon-nucleon force in the nuclear medium. However, one can imagine such a field as being built up by the action of all the nucleons, approximating the A -body problem by A one-body problems.

$$H = \left(\sum_{i=1}^A T_i + \sum_{i=1}^A U_i \right) + \left(\sum_{i=1}^A \sum_{j>i}^A V_{ij} - \sum_{i=1}^A U_i \right) \quad (2.4)$$

$$H = H_0 + H_{res}$$

where H_0 describes the motion of the individual nucleons, independent of each other in the same average field and H_{res} the residual interaction. The independent particle model including the effect of the residual interaction is the basis of the nuclear shell model. The smaller the effect of H_{res} , the better the assumption of an average, independent field becomes. The average potential can be derived using self-consistent Hartree-Fock methods or the Hartree-Fock-Bogoliubov approach, including also the pairing correlations amongst the nucleons.

2.3 Nuclear deformation and the Nilsson model

The assumption of approximately independent motion of the individual nucleons in an average field is the basis of the shell model and most microscopic descriptions of finite nuclei. This average potential is produced by the nucleons themselves and their mutual interaction and, in its most simple form, it is spherical. Starting from the spherical shell model, many nuclear properties can be described based on the characteristics of the two-body nucleon-nucleon interaction and its single-particle energies for nuclei with closed or nearly closed shells. On the other hand, when several valence nucleons are present, the residual nucleon-nucleon interactions can drive the nucleus to a more energetically favored deformed configuration. Using the concept of a deformed shape, one can introduce the idea of independent particles moving in a deformed potential, which, if taken as a deformed harmonic oscillator, is known as the Nilsson model [47]. This model introduces possibilities to describe nuclear collective properties, starting from a microscopic basis.

The nuclear surface $R(\theta, \phi)$ can be expanded in terms of a set of multipole λ deformation coordinates $\alpha_{\lambda\mu}$:

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

where R_0 is the radius of a spherical nucleus with the same volume, λ represents the multipole order and $Y_{\lambda\mu}(\theta, \phi)$ are the spherical harmonics [39].

Restricting the description to quadrupole degrees of freedom $\lambda = 2$, the hamiltonian is written and solved in a five-dimensional configuration space, spanned by the quadrupole deformation coordinates $\alpha_{2\mu}$ where $\mu = -2, -1, 0, 1, 2$.

For axially symmetric shapes, all quadrupole deformation coordinates $\alpha_{2\mu}$ with $\mu \neq 0$ vanish; the quadrupole deformation parameter is defined as $\beta_2 = \alpha_{20}$. If deviations from axial symmetry are included, a second deformation parameter γ is needed. These parameters are defined as (with $\alpha_{21} = \alpha_{2-1} = 0$):

$$\begin{aligned} \alpha_{20} &= \beta_2 \cos \gamma \\ \alpha_{22} &= \alpha_{2-2} = \frac{1}{\sqrt{2}} \beta_2 \sin \gamma \end{aligned} \quad (2.6)$$

The three remaining parameters, related to the Euler angles, define the orientation of the body-fixed frame in three-dimensional space.

The larger the value of β_2 the more deformed the nucleus. Positive and negative β_2 values correspond to prolate (like a rugby) and oblate (like a discus) shapes respectively, while $\beta_2 = 0$ represents the spherical shape. Gamma is defined in degrees where $\gamma = 0^\circ$ and $\gamma = 60^\circ$ correspond to prolate and oblate shapes respectively. Completely triaxial shapes have $\gamma = 30^\circ$. Triaxial shapes are characterized by three axes with different lengths, while shapes with axial symmetry always have two of the three axes of equal length. Both variables are illustrated geometrically in Fig. 2.1 and an example of a potential-energy surface for ^{184}Hg , using such a geometrical representation (with deformation parameter $\epsilon_2 \approx 0.95\beta_2$), is given in Fig. 2.2 [48].

In the Nilsson model, the deformation breaks the $2j+1$ degeneracy of the spherical shell model states and the levels are labeled by the asymptotic quantum numbers $\Omega^\pi[\text{Nn}_z\Lambda]$, where the first quantum number gives the Ω value and the parity π . By definition, $\Omega = \Lambda + \Sigma = \Lambda \pm \frac{1}{2}$, where Λ is the projection of the

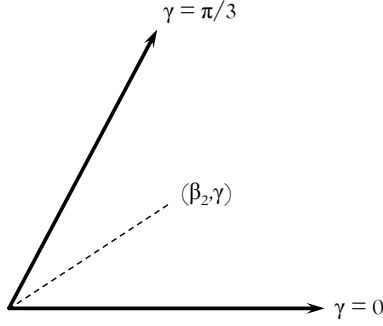


Figure 2.1: Geometric representation of the intrinsic deformation variables β_2 and γ , as defined in the text.

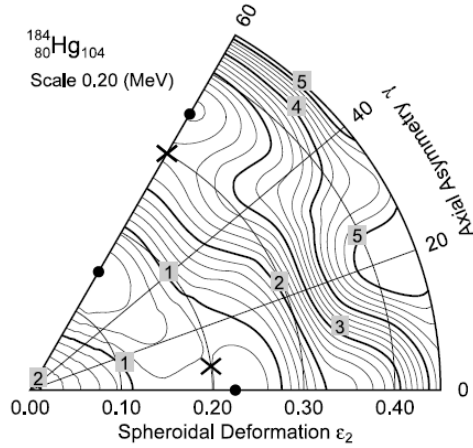


Figure 2.2: Calculated potential-energy surface for ^{184}Hg , indicating two oblate and one prolate minima. Figure is from [48]. Minima are indicated by round dots and saddle points by pairs of crossed lines. The numbers give the energy in MeV of the thicker contour lines that are spaced 1 MeV apart; the spacing between the thinner lines is 0.2 MeV.

angular momentum along the symmetry axis and $\Sigma = \pm \frac{1}{2}$ are the projections of the nucleon spin on the symmetry axis. The quantum number N is the principal quantum number of the major shell and n_z gives the number of nodes in the wavefunction in the direction of the symmetry axis. Each orbit will split according to the Ω value, with slopes depending on the angle $\theta \sim \frac{\Omega}{I}$ and each level can contain only two nucleons. The Nilsson level diagrams as a function of the Nilsson quadrupole deformation parameter ϵ_2 are shown in Fig. 2.3 and 2.4 for protons ($50 \leq Z \leq 82$ and $Z \geq 82$ regions) and Fig. 2.5 for neutrons ($82 \leq N \leq 126$ region) respectively, relevant for the nuclei studied in this work. In these Nilsson diagrams, single-particle energies are plotted in units of the oscillator frequency $\hbar\omega_0 = 41A^{-1/3}$ MeV. The deformation parameter ϵ_2 is related to the previously defined β_2 by

$$\beta_2 = \sqrt{\frac{\pi}{5}} \left(\frac{4}{3}\epsilon_2 + \frac{4}{9}\epsilon_2^2 + \frac{4}{27}\epsilon_2^3 + \dots \right) \quad (2.7)$$

For $\epsilon_2 = 0$, the level structure in the spherical shell model is obtained. Each level is labeled by its respective asymptotic quantum numbers $\Omega^\pi[Nn_z\Lambda]$.

In Fig. 2.4 it is clearly seen that the upward or downward slope of the different proton orbitals gives rise to deformation in the thallium isotopes. For example, between $\epsilon_2 = -0.1$ and -0.2 at the oblate side, the regular $3s_{1/2}(1/2[400])$ orbital has a comparable single-particle excitation energy as the intruder $1h_{9/2}(9/2[505])$ and $1i_{13/2}(13/2[606])$ orbitals. The population of these different single-particle states has indeed been observed experimentally in different thallium isotopes. The isotopes studied in this work are situated around neutron midshell at $N = 104$, indicated in Fig. 2.5 by the bold red line.

2.4 Mean-field description

The mean-field description of the nuclear many-body problem starts from the two-body nucleon-nucleon interaction force. Mean-field descriptions use an unprejudiced, self-consistent determination of the nuclear mean field, whereas in the shell model a standard phenomenological single-particle model is taken. The effective force is tuned to describe global nuclear properties such as charge radii and ground state binding energies throughout the nuclear mass surface and is then used to obtain optimized single-particle states in a self-consistent way according to Hartree-Fock (HF) theory, constraining the nuclear density to exhibit different low-multipole deformations. The resulting energy surface, the projection along the axial quadrupole moment parameter, allows

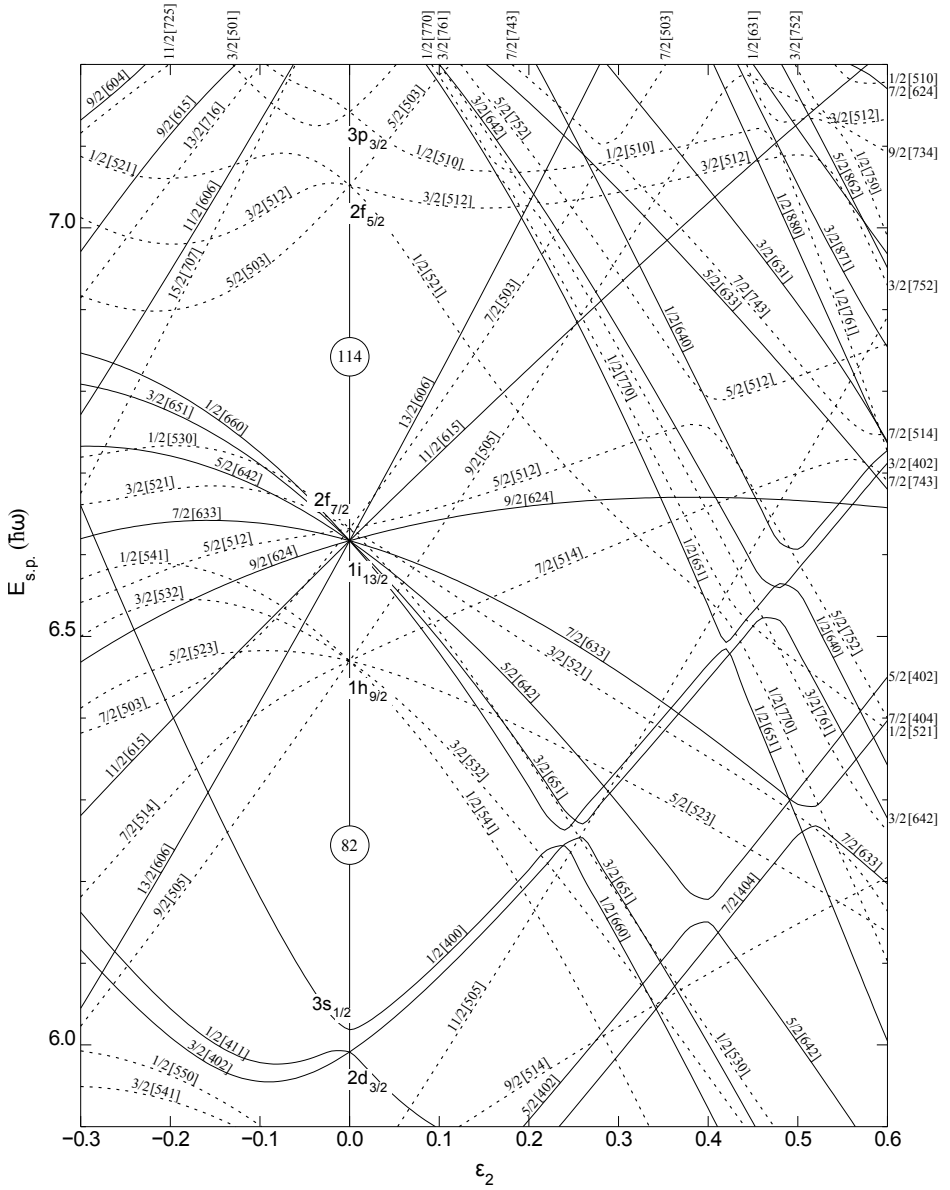


Figure 2.4: Nilsson diagram for protons, $Z \geq 82$ [49].

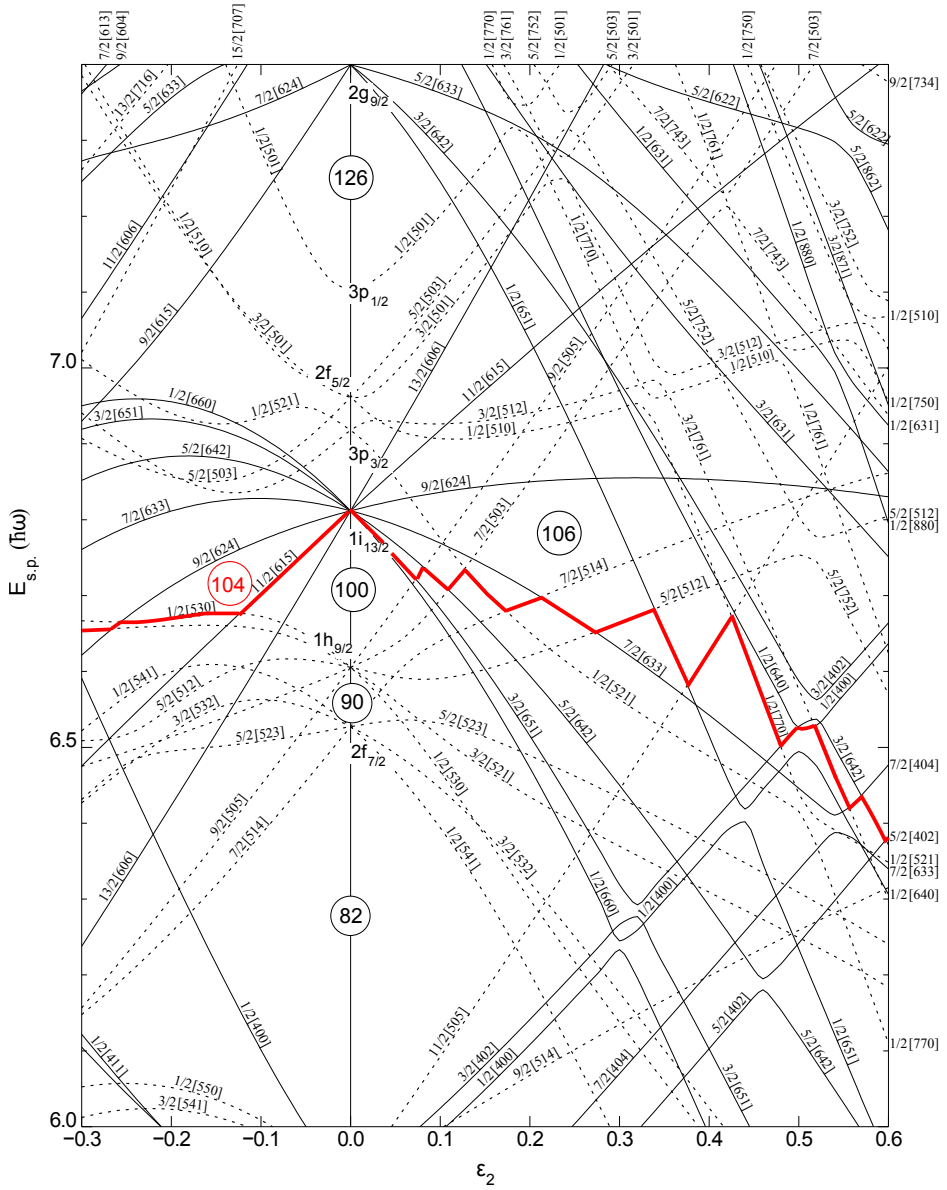


Figure 2.5: Nilsson diagram for neutrons, $82 \leq N \leq 126$ [49]. Neutron midshell, $N = 104$, is indicated with the bold red line.

for the identification of minima that often correspond to a non-spherical density distribution. The HF approximation is inadequate for a description of nuclear properties that are strongly influenced by pairing correlations and in Hartree-Fock-Bogoliubov (HFB) theory, the mean-field concept is generalized to include a pairing field. In the mean-field calculations, symmetries can be broken by the interaction and it is important to restore these to construct states with fixed proton and neutron number, correct isospin and angular momentum. For a detailed overview of self-consistent mean-field models for nuclear structure, we refer to [29].

2.5 Spectroscopic information from α decay

In the region of the neutron-deficient isotopes around $Z = 82$, two main radioactive decay modes occur, namely β^+ /EC decay and α decay. In β^+ /EC decay, the nucleus can correct a proton excess by directly converting a proton into a neutron and emitting a positron (β^+ particle) and electron neutrino (ν_e). Electron capture is a process in which the proton-rich nucleus absorbs an inner atomic electron, thereby changing a nuclear proton to a neutron and simultaneously causing the emission of an electron neutrino. In α decay a nucleus emits a ${}^4\text{He}$ nucleus, also called α particle. These different processes are expressed schematically by the following equations:

$$\beta^+ : {}^A_Z X_N \rightarrow {}^A_{Z-1} X'_{N+1} + e^+ + \nu_e \quad (2.8)$$

$$EC : {}^A_Z X_N + e^- \rightarrow {}^A_{Z-1} X'_{N+1} + \nu_e \quad (2.9)$$

$$\alpha : {}^A_Z X_N \rightarrow {}^{A-4}_{Z-2} X'_{N-2} + \alpha \quad (2.10)$$

In the β^+ /EC decay of a certain nuclear state, different levels in the daughter nucleus can be fed. Further de-excitation to the ground state, possibly through short-living states or to long-living isomeric states proceeds through the emission of γ rays and internal conversion. Alpha decay is subject to a more severe energy dependence and other selection rules compared to β^+ /EC decay. As a consequence, the number of levels fed in α decay is limited, which makes the assignment of the transitions relatively straightforward. In α decay, information on the lowest-lying energy levels is obtained, while in β^+ /EC decay a larger part of the level scheme of the daughter nucleus is populated. Nevertheless, α decay constitutes a valuable spectroscopic method. The selectivity of the

α -decay process renders direct information on the characteristics of the initial and final state of a transition. It should also be noted that the detection of α particles suffers very little from background radiation compared to γ or X-ray radiation.

Predominantly low-lying states are strongly populated due to the strong Q -value dependence of the quantum-mechanical tunneling probability through the Coulomb barrier. The angular momentum dependence of the barrier penetration probability is contained in the centrifugal term through an $\ell(\ell + 1)$ dependence. Alpha decay is also sensitive to the specific orbitals that are involved in the decay. The parity change associated with α emission is given by $(-1)^\ell$, with ℓ the angular momentum carried away by the α particle, indicating which transitions are forbidden or allowed. The formation probability of the α particle in the nucleus, expressed by the reduced α -decay width, is extremely sensitive to the overlap in wave functions between the initial state and final state plus α particle, strongly enhancing $\Delta\ell = 0$ transitions between states of similar origin.

2.5.1 General properties of α decay

The observables from an α -decay experiment are the energy E_α , intensity I_α , half-life $T_{1/2}$ and α -branching ratio b_α .

Conservation of energy in the α -decay process, assuming the initial decaying nucleus X to be at rest, leads to:

$$m_X c^2 = m_{X'} c^2 + T_{X'} + m_\alpha c^2 + T_\alpha \quad (2.11)$$

where mc^2 is the rest energy of the different particles and T represents the kinetic energy of the different final particles.

Defining the Q_α value as the net energy released in the process, given by the difference of the initial and final masses, equation (2.11) can be rewritten as

$$Q_\alpha = T_{X'} + T_\alpha = M_X - M_{X'} \quad (2.12)$$

and, imposing conservation of linear momentum, in a non-relativistic approximation, the kinetic energy of the α particle T_α can be expressed as a function of the Q_α value as

$$T_\alpha = \frac{Q_\alpha}{1 + \frac{m_\alpha}{m_{X'}}} \quad (2.13)$$

This implies that the α particle carries most of the Q_α value. The energy of α particles emitted in the lead region is around 5 - 7 MeV, which leads to a recoil energy of the heavier nucleus X' of about 100-150 keV, which is not negligible.

The transition probability for α decay can be separated into two contributions: an internal contribution which considers the α -decay process in the absence of a Coulomb barrier, the so-called formation probability and an external contribution, which describes the tunneling through the Coulomb barrier, the so-called penetration factor.

The formation probability expresses the probability that starting from the state I_i^π in the parent nucleus, an α particle is formed so that the resulting daughter nucleus is in the I_f^π state. It is clear that the formation probability contains a wealth of spectroscopic information. However, up to present it is not proven that an α particle is actually pre-formed inside a nucleus, but merely that they behave as if they were. The simplest way to form the α particle is by the weakest bound proton and neutron pairs of the parent configuration. Differences in structure and differences in single-particle character will thus hinder the formation of the α particle.

The penetration factor gives the probability that the formed α particles, leave the parent nucleus. The theory from Gamow [50] and Condon and Gurney [51, 52] describes the α -decay process by tunneling through the Coulomb barrier. This theory also explains the strong energy-dependence of the half-life in α decay, since it is much easier for a more energetic α particle to tunnel through the potential barrier. Contrary to the formation probability, the penetration factor can be relatively easy calculated.

The total α -transition probability is the product of the formation probability and the penetration factor. One can thus get information on the formation probability of the α particle from the experimentally determined transition probability and the calculated penetration factor. Following the commonly used description of Rasmussen [53] a measure of the formation probability of an α particle inside the nucleus can be expressed by the reduced α -decay width (δ^2)

$$\delta^2 = \frac{h \ln 2 b_\alpha}{T_{1/2} 100 P} \quad (2.14)$$

where h is Planck's constant and P is the penetration factor. The reduced α -decay width contains essentially all nuclear-structure information and is commonly expressed in keV.

The reduced α -decay width is proportional to the overlap of the wave function of the parent nucleus and that of the daughter nucleus plus the α particle. This overlap does not depend on the energy of the α particle; hence, the energy dependence of the α -decay rate is eliminated in the reduced width and it is

mainly determined by the structure of the initial and final states. For instance, if the initial and final states have a similar structure, spin and parity, the α decay is said to be allowed or unhindered. Typical reduced α -decay widths for such transitions in the mass region studied in this work range between 30 and 70 keV. Transitions which proceed between states with a different structure, spin or parity are hindered and the reduced α -decay widths are much smaller than those of unhindered transitions.

2.6 γ -ray spectroscopy

2.6.1 General properties of γ decay

Gamma-ray decay occurs when a nucleus in an excited state releases its excess energy by emission of electromagnetic radiation, i.e. a photon. We have

$${}^A X^* \rightarrow {}^A X + \gamma \quad (2.15)$$

where the symbol $*$ indicates an excited state of the nucleus. Note that there is no change in Z or A during this type of decay, only the release of energy. These excited states decay through the emission of one or more γ rays to the ground state or an isomeric state. Gamma rays have energies typically in the range of 0.1 to 10 MeV, characteristic of the energy difference between nuclear states.

Conservation of total energy and linear momentum for γ decay (in a non-relativistic framework) leads to

$$E_\gamma = E_i - E_f - T_R \quad (2.16)$$

where E_i, E_f is the energy of the initial excited and final state respectively, E_γ the energy of the emitted γ ray and T_R the kinetic energy of the recoiling nucleus.

This recoil energy is generally negligible since it is usually far smaller than the experimental uncertainty with which we can measure energies. Thus for the remaining part of this work, we will assume $E_\gamma = E_i - E_f = \Delta E$.

The probability per unit time for photon emission, i.e. the decay constant, is given by

$$\lambda(\sigma L) = \frac{P(\sigma L)}{\hbar\omega} = \frac{2(L+1)}{\epsilon_0 \hbar L [(2L+1)!!]^2} \left(\frac{\omega}{c}\right)^{2L+1} [m_{fi}(\sigma L)]^2 \quad (2.17)$$

with $m_{fi}(\sigma L)$ the matrix element of the multipole operator $\hat{m}(\sigma L)$, with $\sigma = E$ or M for an electric or magnetic transition.

$$m_{fi}(\sigma L) = \int \psi_f^* m(\sigma L) \psi_i dv \quad (2.18)$$

where the integral is carried out over the volume of the nucleus. ψ_i and ψ_f represent the initial and final wave functions.

It is clear that $\lambda(\sigma L)$ can be evaluated only if the initial and final wavefunctions ψ_i and ψ_f are known since it is governed by the matrix element of the multipole operator $\hat{m}(\sigma L)$. Victor Weisskopf derived a general expression for the transition probability with the assumption that the transition results from the change of a single particle inside a nucleus with uniform density and using the nuclear radius $R = R_0 A^{1/3}$ [54]. These estimates for the transition rates are known as Weisskopf estimates and provide reasonable relative comparisons of the transition rates. The Weisskopf estimates for the decay constants of the four lowest order multipoles are given in Table 2.1, where E is given in MeV and A is the mass number of the nucleus. From the probabilities in Table 2.1 it emerges that the lower multipolarities are faster and thus dominant and, for a given multipole, the electric transition is about two orders of magnitude more likely than the magnetic one for medium and heavy nuclei. Hence, the most commonly observed transitions are the electric and magnetic dipoles (E1 and M1) and the electric quadrupole (E2) transitions.

In decay spectroscopy studies, the retardation factor F_w of an electromagnetic transition is often determined. The retardation factor is the ratio of the theoretically expected rate, i.e. the Weisskopf estimate, and the observed rate of a transition. When the observed decay rate is much smaller than the Weisskopf estimate, the rate of the transition corresponds to a single-particle transition. On the other hand, if the transition rate is much larger than the Weisskopf estimate one might suspect that more than one single nucleon is involved in the transition. An example of a strongly retarded transition can be found in the odd-odd ^{186}Tl isotope. In ^{186}Tl the characteristic E3 transition from a $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ to a $[\pi 1s_{1/2} \otimes \nu 1i_{13/2}]$ is retarded with a factor of about 6500 relative to the Weisskopf estimate. This very slow E3 transition can be explained by the fact that it is j -forbidden ($\Delta j = 4$) in the framework of the single-particle Weisskopf estimates and is thus highly retarded (see also chapter 4).

A multipole of order L transfers an angular momentum of $L\hbar$ per photon. Conservation of momentum requires that the total initial angular momentum be equal to the final angular momentum. The relative parity of the initial and final levels determines whether the emitted radiation is of the electric or magnetic type. This leads to the following angular momentum and parity selection rules for γ decay:

$$|I_i - I_f| \leq L \leq I_i + I_f \quad (L \neq 0) \quad (2.19)$$

Table 2.1: Weisskopf estimates of decay constants for lowest order multipoles.

$\lambda(\text{EL}) \text{ (s}^{-1}\text{)}$	$\lambda(\text{ML}) \text{ (s}^{-1}\text{)}$
$\lambda(\text{E1}) = 1.0 \times 10^{14} \text{ A}^{2/3} \text{ E}^3$	$\lambda(\text{M1}) = 5.6 \times 10^{13} \text{ E}^3$
$\lambda(\text{E2}) = 7.3 \times 10^7 \text{ A}^{4/3} \text{ E}^5$	$\lambda(\text{M2}) = 3.5 \times 10^7 \text{ A}^{2/3} \text{ E}^5$
$\lambda(\text{E3}) = 34 \text{ A}^2 \text{ E}^7$	$\lambda(\text{M3}) = 16 \text{ A}^{4/3} \text{ E}^7$
$\lambda(\text{E4}) = 1.1 \times 10^{-5} \text{ A}^{8/3} \text{ E}^9$	$\lambda(\text{M4}) = 4.5 \times 10^{-6} \text{ A}^2 \text{ E}^9$

$$\pi_E = (-1)^L \quad \pi_M = (-1)^{L+1} \quad (2.20)$$

The exception to the angular momentum selection rule occurs when $I_i = I_f = 0$ because there are no monopole ($L = 0$) transitions in which a single photon is emitted. In this case, the decay occurs through internal conversion, which is discussed in the next section.

2.6.2 Internal conversion

Internal conversion is an electromagnetic process which competes with γ emission. The electromagnetic multipole fields of the nucleus interact with the atomic electrons and can cause one of the existing electrons to be emitted from the atom. The kinetic energy T_e of the emitted electron equals the transition energy ΔE , less the electron binding energy B that must be supplied to knock the electron loose from its atomic shell:

$$T_e = \Delta E - B \quad (2.21)$$

Equation (2.21) indicates that the internal conversion process has a threshold energy equal to the electron binding energy in a particular shell. Conversion electrons are thus labeled according to the electronic shell from which they are emitted, i.e. K, L, M, $\dots$. Following the emission of an electron, the so created vacancy is filled by electrons from higher shells and the emission of characteristic X-rays is observed in this process.

In some cases internal conversion is strongly favored over γ emission, in others it is the opposite. An internal conversion coefficient α_{CE} is defined as

$$\alpha_{CE} = \frac{\lambda_e}{\lambda_\gamma} = \frac{I_e}{I_\gamma} \quad (2.22)$$

where $\lambda_{e,\gamma}$ represents the decay probability arising from internal conversion or γ emission respectively and I their respective intensities. The total decay probability λ_t is given by

$$\lambda_t = \lambda_\gamma + \lambda_e = \lambda_\gamma(1 + \alpha_{CE}) \quad (2.23)$$

where α_{CE} is the total internal conversion coefficient, defined as the sum of the partial coefficients:

$$\alpha_{CE} = \alpha_K + \alpha_L + \alpha_M + \dots \quad (2.24)$$

where α_i corresponds to the partial coefficient representing atomic shell i .

The process of internal conversion is electromagnetic in origin and has thus a similar matrix element to that of equation (2.18). The difference comes from the fact that for internal conversion, the initial state includes a bound electron and the final wave function includes a free-particle wave function. The conversion coefficient thus depends on the atomic number of the atom in which the process occurs and on the energy of the transition and its multipolarity. The multipolarity of an electromagnetic transition can then be deduced from the experimental determination of the conversion coefficient. The theoretical conversion coefficients used in this work come from Ref. [55].

2.7 Laser spectroscopy

Laser spectroscopy of radioactive nuclei combines aspects of nuclear, atomic and laser physics. Many experimental techniques study the properties of nuclear decay, for example the energy of α or β particles or γ rays. In this way, nuclear structure is studied by observing the changes in going from one nuclear state to another. Optical spectroscopy is an alternative approach, offering a probe of the nuclear states themselves.

In laser-spectroscopy studies, high-resolution laser frequency scans are performed over a selected transition between atomic levels in order to observe a resonance pattern. These laser spectroscopy scans provide nuclear model-independent measurements of the nuclear spin, magnetic dipole moment, spectroscopic quadrupole moment and differences in mean-square charge radius through

the hyperfine splitting (HFS) and isotope shift. The HFS and isotope shift result from the interaction between the nuclear charge distribution and the electromagnetic field of the atomic electrons and affect the atomic spectrum. The interaction is purely electromagnetic and a common approach is to expand the electromagnetic transition operator in a multipole series. The electric monopole corresponds to the nuclear charge and gives rise to an overall shift in the electronic fine-structure levels, i.e. the isotope shift. The magnetic monopole vanishes, since no analogue of the electrical charge is known to exist for the magnetic case. As the nuclear matrix elements are diagonal for states with a well defined J^π , only the even parity moments are contributing. The magnetic dipole and the electric quadrupole moment give rise to a splitting of the atomic levels, i.e. the HFS [41].

2.7.1 Hyperfine structure

If the atomic spin, J , and nuclear spin, I , are both greater than zero, then the level is split into hyperfine levels. The coupling of I and J defines the quantum number F , where F ranges from $|I - J|$ to $I + J$. The hyperfine interaction removes the degeneracy, splitting the different F -levels. The perturbation of each level is given by [56]:

$$\frac{\Delta E}{h} = \frac{K}{2}A + \frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)}B \quad (2.25)$$

where $K = F(F+1) - I(I+1) - J(J+1)$. A and B are the magnetic dipole hyperfine coupling constant and electric quadrupole hyperfine coupling constant respectively and are expressed as:

$$A = \frac{\mu_I B_e(0)}{IJ} \quad (2.26)$$

$$B = eQ_s \left\langle \frac{\partial^2 V_e}{\partial z^2} \right\rangle \quad (2.27)$$

The first term in equation (2.25) accounts for the interaction between the magnetic dipole moment of the nucleus, μ_I , and the magnetic field created by the electrons at the nucleus, $B_e(0)$. The second term arises when both $I > 1/2$ and $J > 1/2$ as a result of the nuclear spectroscopic quadrupole moment, Q_s ¹, interacting with the electric field gradient, $\langle \frac{\partial^2 V_e}{\partial z^2} \rangle$, produced by the electrons.

¹The spectroscopic quadrupole moment is different from the intrinsic quadrupole moment Q_0 , which is induced by the non-spherical charge distribution of the nucleus and which is defined in the intrinsic frame of the nucleus [41].

The magnetic dipole moment is calculated as the expectation value of the z component (μ_z) of the vector magnetic dipole operator $\vec{\mu} = g_\ell \vec{\ell} + g_s \vec{s}$, in the state $M = I$:

$$\mu(I) = \langle I, M = I | \mu_z | I, M = I \rangle \quad (2.28)$$

The gyromagnetic factors are given by $g_s = 5.58$, $g_\ell = 1$ for protons; $g_s = -3.82$, $g_\ell = 0$ for neutrons.

In analogy to the magnetic dipole moment the electric quadrupole moment is defined as

$$Q_s(I) = \langle I, M = I | Q_z | I, M = I \rangle \quad (2.29)$$

with Q_z the z-component of the quadrupole operator, defined in terms of spherical harmonics as

$$Q = \sqrt{\frac{16\pi}{5}} \sum_{k=1}^A e(k) r_k^2 Y_2^0(k) \quad (2.30)$$

The nuclear moments can also be calibrated using the known moments of another isotope of the element under study, via the ratios

$$\mu = \mu_{ref} \frac{IA}{I_{ref}A_{ref}} (1 + \Delta) \quad (2.31)$$

$$Q_s = Q_{s,ref} \frac{B}{B_{ref}} \quad (2.32)$$

where Δ is the hyperfine anomaly. The hyperfine anomaly arises from differences in charge and magnetization distribution within the nucleus.

In case of odd-odd nuclei, the magnetic moment may be calculated by taking into account the separate contribution from the odd-proton (μ_π) and the odd-neutron (μ_ν) [37]:

$$\mu = \frac{I}{2} \left[\frac{\mu_\pi}{I_\pi} + \frac{\mu_\nu}{I_\nu} + \left(\frac{\mu_\pi}{I_\pi} - \frac{\mu_\nu}{I_\nu} \right) \frac{I_\pi(I_\pi + 1) - I_\nu(I_\nu + 1)}{I(I + 1)} \right] \quad (2.33)$$

where I_π and I_ν are the spins of the odd-proton and odd-neutron states respectively.

2.7.2 The mean-square charge radius

The radius of a nucleus cannot be determined exactly since the boundaries of a nucleus are not well defined. The average matter distribution is such that there is an inner region of roughly constant density surrounded by a diffuse region where the density gradually falls to zero. As mentioned in section 2.1, one can distinguish between the matter radius, determined by all nucleons, and the charge radius, composed by only the protons of the nucleus. The mean-square charge radius, which is the average of the spread of the nuclear charge distribution, is defined as [39]

$$\langle r^2 \rangle = \frac{\int \rho(r) r^2 dV}{\int \rho(r) dV} \quad (2.34)$$

where $\rho(r)$ represents the distribution of the charge density of the protons in the nucleus.

The dominant part of $\langle r^2 \rangle$ is described by the previously introduced droplet model [44, 45, 57]. The mean-square charge radii are then expressed by:

$$\langle r^2 \rangle = \langle r^2 \rangle_{DM} \left(1 + \frac{5}{4\pi} \langle \beta^2 \rangle \right) \quad (2.35)$$

where $\langle \beta^2 \rangle$ represents the mean-square deformation and $\langle r^2 \rangle_{DM}$ is the droplet-model prediction for a spherical nucleus with the same A and Z values. In this work, the second set of parameters from Ref. [57] is used to calculate the droplet-model predictions. It should be noted that in equation (2.35) only the quadrupole deformation β_2 is taken into account and higher order deformations ($\beta_3, \beta_4, \dots$) are neglected since their contribution is usually small.

2.7.3 Isotope shift

The displacement of the hyperfine structure centroid from one isotope to another is known as the isotope shift. To a very good approximation [58], this can be written as the sum of the mass shift and field shift terms:

$$\delta\nu_{IS}^{A,A'} = \delta\nu_{MS}^{A,A'} + \delta\nu_{FS}^{A,A'} \quad (2.36)$$

where $\delta\nu^{A,A'} = \nu^{A'} - \nu^A$

The mass shift is an effect of the finite mass of the nucleus. It is in turn a combination of two terms: the normal mass shift ($\delta\nu_{normal}$) is caused by the

change in the reduced mass of the isotope, while the specific mass shift relates to the influence of the correlated electronic momenta on the nuclear motion. The normal mass shift is given by

$$\delta\nu_{normal}^{A,A'} = \frac{m_e}{m_p} \frac{A - A'}{AA'} \nu^A \quad (2.37)$$

where ν^A is the transition frequency, m_e and m_p are the electron and proton masses, respectively. As the specific mass shift has the same A -dependence, the total mass shift can be written as:

$$\delta\nu_M^{A,A'} = \delta\nu_{NMS}^{A,A'} + \delta\nu_{SMS}^{A,A'} = M \frac{A - A'}{AA'} \quad (2.38)$$

where the mass factor, M , including both normal (NMS) and specific mass (SMS) shift, is atomic transition dependent. For light nuclei, the mass shift gives an appreciable contribution, but for heavier nuclei, the field shift is dominant. The field shift arises from the change in the electrostatic potential felt by the electrons due to the difference in the spatial extent of the nuclear charge distribution. It can be split into a nuclear factor $\lambda^{A,A'}$ and an electronic factor F , which depends on the electron density at the nucleus.

The isotope shift can therefore be expressed as:

$$\delta\nu^{A,A'} = M \frac{A - A'}{AA'} + F \lambda^{A,A'} \quad (2.39)$$

where $\lambda^{A,A'} = K(Z) \delta \langle r^2 \rangle^{A,A'}$ and $K(Z)$ takes the contribution of higher-order radii moments in $\lambda^{A,A'}$ into account (see [59, 60]). The nuclear properties are contained within $(A - A')/AA'$ and $\delta \langle r^2 \rangle^{A,A'}$, and the atomic factors, F and M , contain the optical transition dependence.

3 | Experimental setup

The decay and optical spectroscopy experiments (IS466 in July 2010 and IS511 July 2011) described in this thesis were performed at the radioactive ion-beam facility ISOLDE at CERN (Geneva, Switzerland) [33]. This facility can deliver pure, low-energy beams of radioactive nuclei with half-lives down to milliseconds. This chapter provides information about the beam production, the detection setup that was used and the data acquisition system.

3.1 The ISOLDE facility

The experiments were carried out at the CERN Isotope Separator On-Line Device (ISOLDE) facility [33]. The radioactive thallium nuclei were produced via spallation reactions in a thick 50 g/cm^2 UC_x target, irradiated with a proton beam from the Proton Synchrotron Booster (PSB) at an energy of 1.4 GeV and an intensity up to $2.1 \mu\text{A}$. The proton beam delivered by the PSB consisted of a repeated sequence of pulses (typically 35-40) separated by periods of 1.2 s, called a supercycle. After proton impact, recoiling nuclei stopped in the target matrix, diffused out and effused towards the hot cavity via the transfer line, both kept at a high temperature of $\sim 2300 \text{ K}$. In the hot cavity, the thallium atoms were then ionized through different ionization mechanisms.

The heart of an on-line isotope separator is its target and ion source. The target should assure a **fast** liberation of the radioactive nuclei produced in **large** amounts of target material, which sometimes are conflicting requirements. The combination with the ion source should be able to produce efficiently an ion beam which preferably should only contain isotopes from one chemical element. The intensity and physical composition of the ion beams produced at ISOL facilities are strongly dependent on the type of source used to ionize the isotope of interest. There are three different types of ion sources in use at ISOLDE:

surface ion sources, plasma ion sources and laser ion sources. The data in this work have been obtained using a surface ion source and a laser ion source. The surface ion source is the simplest setup for ionizing atoms produced in the target. The ionizer consists only of a metal tube (“line”), for example tantalum or tungsten, which has a higher work function than the ionization potential of the atom that should be ionized. Surface ionization can be used efficiently for thallium isotopes since the ionization potential (6.1 eV), is smaller than the work function of the tube (for tungsten and tantalum about 8 eV) . The ratio between the ion density (n_i) and the neutral density (n_0) of a certain element with ionization potential (E_i) at a heated surface, with temperature (T) and work function (ϕ) is given by the Langmuir equation[61]:

$$\frac{n_i^+}{n_0} = \frac{g_i^+}{g_0} \exp [(\phi - E_i)/kT] \quad (3.1)$$

where g_i and g_0 is the statistical weight of the ionic and atomic ground state, respectively. The ionization efficiency is given by

$$\epsilon_{ion} = \frac{n_i^+}{n_i^+ + n_0} \quad (3.2)$$

Although thallium isotopes are easily surface ionized, the total ionization efficiency can be increased through resonant laser ionization and, as will be shown later, this also enables enhanced isomer content in the final ion beam. The resonance ionization laser ion source (RILIS) [30, 31, 32] at ISOLDE is based on the method of laser step-wise resonance ionization of atoms in a hot metal cavity. The RILIS is a system of pulsed laser beams which interact with the atomic vapor present in the tube that is kept at high temperature. Each laser wavelength is precisely tuned so that the photon energies match the unique combination of successive electronic transition energies within an ionization scheme for a specific element. By exploiting the unique electronic structure of different atomic species the RILIS provides in this way a rapid, efficient and highly Z selective ionization process. A schematic representation of radioactive ion-beam production at the ISOLDE facility is given in Fig. 3.1.

The target-ion source is kept at an electrostatic potential of 40 kV relative to the ground potential. This allows the newly formed ions to be extracted and accelerated to a kinetic energy of 40 keV. Before transportation to the detection setup, the beam is sent to one of the two magnetic mass separators for separation according to the mass-to-charge ratio. The smaller of the two, the General Purpose Separator (GPS) has one bending magnet and a mass resolving power of more than 1000 [63]. The High Resolution Separator (HRS) consists of two bending magnets with an elaborate ion-optical system for higher order

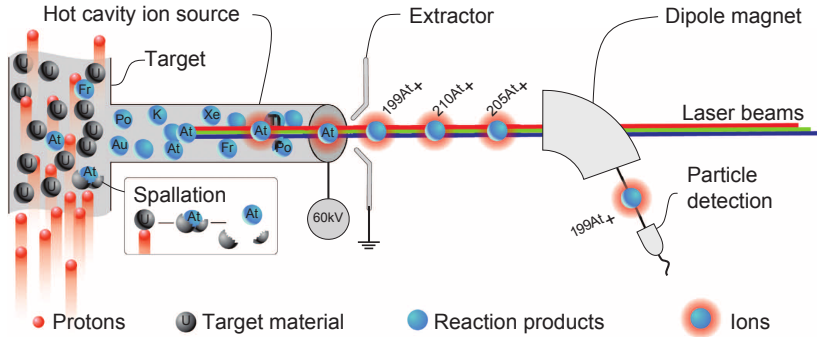


Figure 3.1: [Color] Schematic overview of radioactive ion-beam production at the ISOLDE facility. Different isotopes of various chemical elements are produced by protons impinging on a thick target. After selective ionization, extraction and mass separation, pure isotopic ion beams are transported towards the detection setup. Here the lasers are tuned to ionize astatine and the mass separator is set to ^{199}At . Figure from [62].

corrections. Its mass resolving power exceeds 5000 [63]. After mass separation, the ion beam enters the detection setup. A (partial) layout of the ISOLDE facility is given in Fig. 3.2. The position of the detection setup used to collect the data in this work, the Windmill system, is also indicated. During the IS466 experimental campaign, HRS was used, while the data from IS511 were collected with GPS.

3.2 Detection setup

The data in this work have been obtained with the so-called Windmill (WM) detection system [34, 35, 36]. The WM is a vacuum chamber with a rotating wheel that hosts ten carbon foils (6 mm diameter, $20 \mu\text{g}/\text{cm}^2$ thickness [64]). After a measurement the wheel can be rotated to move away the radioactivity from the implantation position and present a fresh foil in front of the ion beam. For the data collected in this work, this was typically after one supercycle. To detect the decay products, four silicon (Si) and two single crystal high-purity germanium (HPGe) detectors were installed surrounding the carbon foils. A schematic representation of the WM system is given Fig. 3.3. Two Si detectors were placed at the implantation position in close geometry, covering a solid angle of 24% of 4π . One Si detector (Si1) was placed upstream. The beam

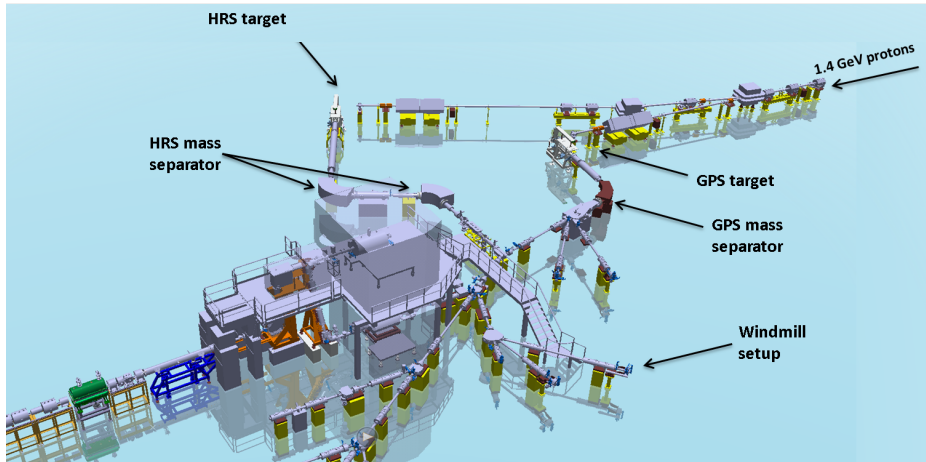


Figure 3.2: [Color] Layout of the CERN ISOLDE facility.

passed through a hole of 8 mm diameter. The other Si detector (Si2) was placed downstream behind the implantation foil. Longer-lived radiation was detected at the decay position with two Si detectors placed in a similar manner as the implantation detectors (Si3 placed downstream; Si4 placed upstream). Meanwhile, implantation and measurements continued on a fresh foil in front of the ion beam. The energy resolution (full width half maximum, FWHM) of these detectors for α decays in the range of 5000-7000 keV was ~ 25 -30 keV for ^{184}Tl (IS511) and ~ 35 keV for ^{182}Tl (IS466). The higher FWHM during the ^{182}Tl campaign is due to the combination of the fact that this run was dedicated to β -delayed fission (βDF) measurements, which require an energy range of the silicon detectors up to 100 MeV and the influence of electronic noise. The absolute efficiency for the Si detectors was determined with two ^{241}Am sources mounted on the wheel of the WM and making use of α - γ coincidences between the 5485 keV ^{241}Am α decay and the 60 keV γ ray. The Si detectors were also used for detecting β^+ particles and conversion electrons (CE). The detection efficiency of Si2 for electrons was determined by using the CE from the 375 keV E0 transition ($0_2^+ \rightarrow 0_1^+$) in ^{184}Hg , populated in the β^+ /EC decay of ^{184}Tl . Due to the limited resolution of the Si2 detector, M(and higher order)-CE could not be resolved from L-CE. This results in 7.6(16)% at 292 keV (K conversion) and 7.2(17)% at 360 keV (L,M conversion) (see [65]). A more detailed description of the determination of the detection efficiency for α -particles and electrons can be found in appendices A and B, respectively.

To detect γ rays, two HPGe detectors were placed outside the vacuum chamber, downstream at 0° (Ge1, 90% relative efficiency, ~ 1 cm from the foil) and at 90°

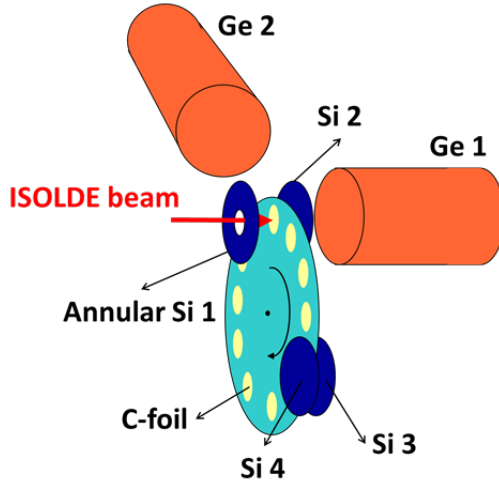


Figure 3.3: [Color] Schematic representation of the WM detection system. The position of the four Si and two Ge detectors is indicated with respect to the incoming beam.

(Ge2, 70% relative efficiency, ~ 3 cm from the foil) with respect to the direction of the incoming beam. The typical energy resolution (FWHM) of each crystal for 1.3 MeV γ radiation was ~ 3.1 keV. Energy and efficiency calibrations were performed using standard calibrated sources of ^{133}Ba , ^{137}Cs , ^{60}Co , ^{241}Am and ^{152}Eu . The absolute photo-peak efficiency for the 506 keV line, the strongest transition in the internal transition (IT) decay of the (10^-) state in ^{184}Tl , was 4.9(1)% and 0.76(3)% in Ge1 and Ge2 respectively, for the given geometry. The obtained efficiency curves for Ge1 and Ge2 are shown in Fig. 3.4 and Fig. 3.5 respectively. For the details on the determination and fitting procedure of the efficiency curves with the standard calibrated sources, we refer to [66].

Due to the combination of a high γ -multiplicity in the β decay of ^{182}Tl and ^{184}Tl and close source-to-detector configuration, the effective efficiency for the HPGe detectors was lower than that determined by the efficiency calibration using standard calibrated sources. To account for this effect, the efficiency for the 351 and 367 keV γ lines, the $2^+ \rightarrow 0^+$ transition in the β -decay daughters ^{182}Hg and ^{184}Hg respectively, has been determined by γ - γ coincidences. With this method, an efficiency for the 351 keV line in ^{182}Hg of 0.77(7)% and 0.44(4)% was obtained for Ge1 and Ge2 respectively. For the 367 keV line in ^{184}Hg , efficiencies of 2.0(2)% and 0.71(7)% were obtained for Ge1 and Ge2 respectively. The difference in effective efficiency is due to the higher multiplicity in the β decay

of ^{182}Tl compared to ^{184}Tl . A more detailed description of the determination of the detection efficiency for γ rays using γ - γ coincidences can be found in appendix C.

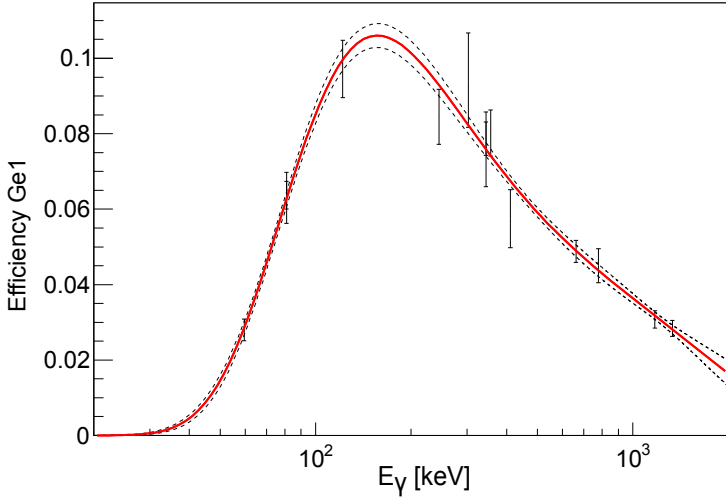


Figure 3.4: [Color] Efficiency curve for Ge1 (red). The error on this fitted curve is given by the dashed lines.

An overview of the different detectors used during each experimental campaign is given in Table 3.1.

3.3 Data acquisition

In the detection setup, digital electronics (digital gamma finder (DGF) modules [67]) were used to acquire the data. The DGF family of digital pulse processors features unique capabilities for measuring both the amplitude and shape of pulses in nuclear spectroscopy applications. Time as well as energy information are stored for every event in one of the detectors. Unfortunately, due to the high count rates, we suffered from a dead time effect caused by the buffer read-outs of the DGF modules. These dead time effects can be assessed by the use of a 10 Hz pulser to indicate the effective measurement time. Dead time effects can greatly influence the determination of the half-life of an isotope or isomer. In chapter 4 this effect is discussed in more detail.

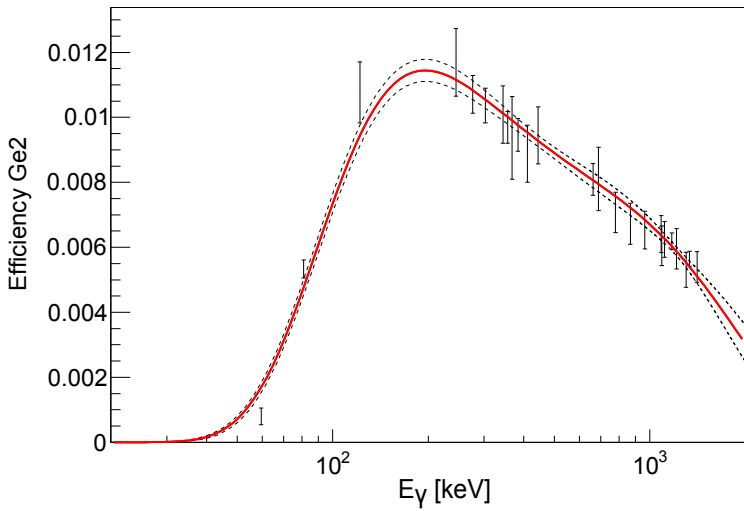


Figure 3.5: [Color] Efficiency curve for Ge2 (red). The error on this fitted curve is given by the dashed lines.

Four reference timing signals are recorded in addition to the pulser and timing and energy information from each detector. These signals are the arrival of a proton pulse, the start of a supercycle, the start of the acquisition system and the closing of the macro beam gate. These so-called timestamps are useful for various reasons, for example to discriminate between radiation arising from short- and long-living radioactive isotopes and for half-life determination. This will be explained in more detail in chapter 4.

In Fig. 3.6 a schematic representation of a typical measurement cycle using the WM setup is depicted. The PSB produces proton pulses every 1.2 s and a number of these pulses are sent to ISOLDE, in this example 17 out of the 34 proton pulses per supercycle. The data acquisition is triggered by the start of a new SC, however, it is delayed by a fixed amount of time to coincide with the first PP that arrives at ISOLDE. In this example the implantation time is set to a micro cycle of only a few tens of milliseconds and the macro beam gate is set to coincide with the data acquisition. After each measurement cycle, the wheel of the WM turns and the whole cycle starts again.

Table 3.1: Overview of the different detectors used during each experimental campaign. Information on the corresponding position of each detector can be found in Fig. A.1 and is given in the text.

Experiment	Detector	Type	Serial Number
IS466	Si1	No detector	
	Si2	PIPS ^a , Canberra	74257
	Si3	SB ^b , Ortec	47-038D
	Si4	PIPS ^a , Canberra	74256
	Ge1	HPGe, Canberra	GC9019
	Ge2	HPGe, Canberra	GC7023
IS511	Si1	SB ^b , Ortec	43-051/F
	Si2	SB ^b , Ortec	47-038D
	Si3	PIPS ^a , Canberra	74256
	Si4	PIPS ^a , Canberra	74257
	Ge1	HPGe, Canberra	GC9019, b90008
	Ge2	HPGe, Canberra	GC7023, B88045

^a Passivated Implanted Planar Silicon (PIPS)

^b Surface Barrier (SB)

3.4 Resonant laser ionization of thallium

The two-step resonant ionization scheme for thallium, used in the experimental campaigns when the data presented in this work were collected, is shown in Fig. 3.7. The excitation from the atomic ground state $6p\ ^2P_{1/2}$ to an intermediate electronic state $6d\ ^2D_{3/2}$ (36117.9 cm^{-1}) was performed by a frequency-doubled narrow-band tunable dye laser beam at 276.9 nm with a linewidth of 0.8 GHz. The first step was used for scanning the hyperfine structure. The subsequent ionization of excited thallium atoms was accomplished by a powerful 532 nm beam of 10 kHz EdgeWave Nd:YAG laser, that was also used as a pump source for the first excitation step laser (for details of RILIS laser setup see [31]).

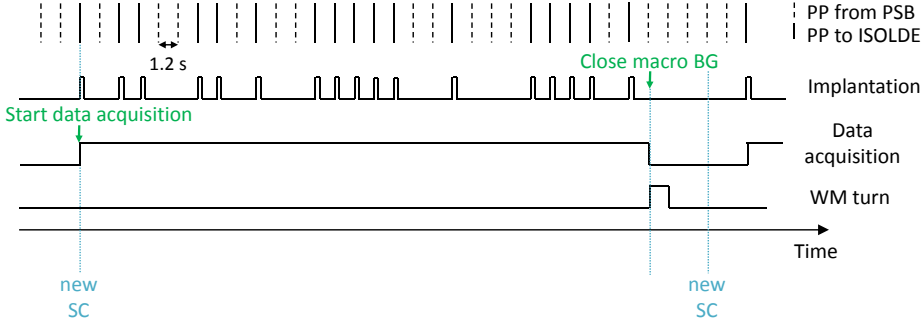


Figure 3.6: [Color] Example of a typical measurement cycle using the WM setup. The PSB produces proton pulses every 1.2 s and a number of these pulses are sent to ISOLDE, here 17 out of the 34 proton pulses per supercycle. The data acquisition is triggered by the start of a new SC, however, it is delayed by a fixed amount of time to coincide with the first PP that arrives at ISOLDE. In this example the implantation time is set to a micro cycle of only a few tens of milliseconds and the macro beam gate is set to coincide with the data acquisition. After each measurement cycle, the wheel of the WM turns and the whole cycle starts again.

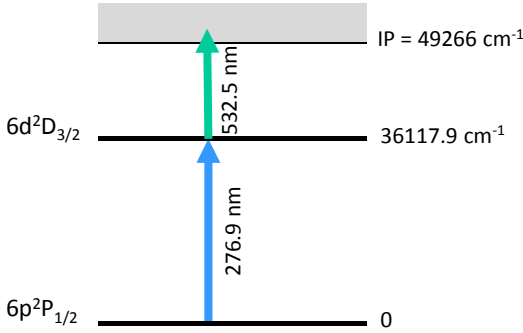


Figure 3.7: [Color] Resonant laser ionization for thallium

4 | Decay spectroscopy of $^{182,184}\text{Tl}$

This chapter presents the results of the laser-assisted decay spectroscopy study of $^{182,184}\text{Tl}$ at the ISOLDE facility using the Resonant Ionization Laser Ion Source. This dual approach was essential to identify the different decay branches of the long-lived states in both isotopes and disentangle the decay schemes. Since thallium has one proton missing in the major proton shell ($Z = 81$) and the neutron number is close to mid-shell ($N = 104$), the interpretation of the results is based on the single-particle properties on one hand and on shape coexistence phenomena on the other hand. This interplay is directly reflected in the properties of both ground and low-lying intruder states. This research has been presented in two peer-reviewed articles [68, 69]:

Paper I : C. Van Beveren et al., "Internal decay of the (10^-) intruder state in ^{184}Tl ", *Physical Review C*, vol. 92, p. 014325, 2015.

Paper II : C. Van Beveren et al., " α -decay study of $^{182,184}\text{Tl}$ ", *Journal of Physics G: Nuclear and Particle Physics* **43**, 025102 (2016)

Both publications are also given in sections 4.1 and 4.3 with slight modifications to their lay-out in order to match the style of this thesis. My main contribution to these papers is the analysis and interpretation of the experimental results, based on fruitful discussions with several co-authors. I was also the corresponding author, responsible for drafting and submitting the articles.

4.1 Internal decay characteristics of ¹⁸⁴Tl (Paper I)

PHYSICAL REVIEW C **92**, 014325 (2015)

Internal decay of the (10⁻) intruder state in ¹⁸⁴Tl

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Abstract

Decay spectroscopy of ¹⁸⁴Tl has been performed at the CERN Isotope Separator On-Line (ISOLDE) facility. An excitation energy of 506.1(1) keV and a half-life of 47.1(7) ms of the intruder based (10⁻) state have been extracted. The internal decay characteristics of this state are determined and discussed, extending the systematics of such states in the even-mass thallium nuclei below neutron mid-shell at N = 104. The retardation factors of the

isomeric M2 and E3 transitions are deduced and compared with retardation factors in neighboring odd-mass and even-mass thallium isotopes. The new information is combined with a review of hindered and unhindered α -decay data of $^{187-192}\text{Bi}$ populating levels in daughter nuclei $^{183-188}\text{Tl}$ and supports the interpretation of the intruder character of the (10^-) state in ^{184}Tl .

4.1.1 Introduction

The neutron-deficient thallium isotopes with one proton less than the $Z = 82$ shell closure, represent an interesting region of the nuclear chart to study in detail shape coexistence [1, 2, 3] in nuclei, i.e. the existence of states with different shapes at low excitation energy in the same atomic nucleus. Extensive investigation of this phenomenon for the lead region together with a substantial development of new theoretical approaches constitutes the basis to further extend the experimental research to the neutron-deficient thallium isotopic chain. For an overview on the experimental evidence and the recent progress in theoretical descriptions, we refer to [4].

In the neutron-deficient, odd-mass thallium nuclei, states with different shapes, whose structure has been linked to specific proton orbitals above and below $Z = 82$, are present at low energy. Next to spherical proton-hole configurations, involving the “normal” $\pi 3s_{1/2}$ and $\pi 2d_{3/2}$ orbitals, one particle-two hole (1p-2h) configurations, involving the $\pi 1h_{9/2}$ and $\pi 1i_{13/2}$ orbitals from above the $Z = 82$ shell closure, that lead to deformation, are identified. Rotational band structures built on top of the latter, so-called intruder states, were observed (see Fig. 18 in [4]). The energy of the 1p-2h band heads show typical parabolic behavior as a function of neutron number, reaching a minimum at $N = 108$, close to $N = 104$, mid-shell between $N = 82$ and $N = 126$. This leads for the case of the $(9/2^-)$ state to isomerism. For $^{185,191,193,195,197}\text{Tl}$, the mean-square charge radii of the $(1/2^+)$ ground state as well as the $(9/2^-)$ isomer were measured. A large isomer shift was observed, implying a larger deformation of the $(9/2^-)$ isomer relative to that of the $(1/2^+)$ ground state [7].

The well-established occurrence of intruder states and shape coexistence in odd-mass thallium isotopes, raises the question of where such states appear in the even-mass thallium isotopes, especially in the lightest isotopes below $N = 104$, and what role the unpaired neutron plays when coupled to the unpaired proton. The coupling of this unpaired neutron and unpaired proton (normal as well as intruder) results in multiplets of states from which some members can become isomeric. This is further complicated by a relatively small energy spacing between these multiplet states and/or the collective bands built on top of the intruder configurations (see e.g. [12]). This results in complex decay

schemes and low-energy, highly converted transitions must be reliably identified and located in the decay schemes.

However, by focusing on isomeric γ radiation and/or by using the selectivity of the α -decay process, the position of the intruder $(8,9,10^-)$ state, originating from the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ coupling, relative to the normal (7^+) state, originating from the $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ coupling, could be measured in a number of thallium isotopes. Also here the energy difference between the intruder and normal configuration exhibits a parabolic behavior; similar to the one observed in the odd-mass thallium nuclei (see Fig. 7b in [70]). It was also found that in the odd-mass as well as the even-mass thallium isotopes, the internal transition (IT) of the intruder isomer is hindered, another fingerprint of shape coexistence [1].

In this paper we focus on the IT of the (10^-) state in ¹⁸⁴Tl. Prior to our study, the location of this isomeric (10^-) state was found at 500(5) keV above the β -decaying (7^+) state, determined through the α decay of ¹⁸⁸Bi, but its half-life and decay characteristics were unknown [71]. The presumed $(10^-)[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ state in ^{188m1}Bi decays by unhindered α decay to the (10^-) state, explained as the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ intruder state in ¹⁸⁴Tl, and by a strongly hindered decay to the (7^+) state, explained as the normal $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ state in ¹⁸⁴Tl. The $(3^+)[\pi 1h_{9/2} \otimes \nu 3p_{3/2}]$ state ^{188m2}Bi decays unhindered to a (3^+) state, explained as the $[\pi 1h_{9/2} \otimes \nu 3p_{3/2}]$ intruder state, 99 keV above the weakly fed (2^-) β -decaying state, explained as the normal $[\pi 3s_{1/2} \otimes \nu 3p_{3/2}]$ state. As the relative position of the α -decaying states in ¹⁸⁸Bi is unknown, the relative position of the (7^+) and (2^-) ¹⁸⁴Tl β -decaying states is not fixed [71]. Direct mass measurements in a Penning trap could not resolve this [72].

In our study, combining the high selectivity of the Resonant Ionization Laser Ion Source (RILIS) [30, 31, 32] with decay spectroscopy and the in-source laser spectroscopy method [73, 74, 75], exotic thallium isotopes down to $N = 97$ have been studied at the Isotope Separator On-Line Device ISOLDE [33] CERN, Geneva, Switzerland, using the Windmill detection system [34, 35, 36]. Complementary decay data on isomerically purified sources were collected. This has led to the characterization of the decay of the (10^-) , (2^-) and (7^+) states in ¹⁸⁴Tl. In the present study, the IT characteristics of the (10^-) intruder state are presented and discussed. The results of the β^+/EC decay of the (7^+) and (2^-) states as well as the α decay of the three long living states will be reported elsewhere, respectively [65] and [69].

4.1.2 Experimental setup

The experiment was carried out at the CERN Isotope Separator On-Line Device (ISOLDE) facility [33]. Protons from the CERN Proton Synchrotron Booster,

with an energy of 1.4 GeV and an intensity up to 2.1 μA , impinged on a 50 g/cm² UC_x target. The proton beam consisted of a repeated sequence of pulses (typically 35-40) separated by periods of 1.2 s, called a supercycle. The radioactive thallium isotopes were predominantly formed through proton-induced spallation. After proton impact, recoiling nuclei stopped in the target matrix, diffused out and effused towards the hot cavity kept at a high temperature of ~ 2300 K. In the hot cavity thallium isotopes were selectively laser-ionized using the same scheme as in the study of [35]. The excitation from the atomic ground state to an intermediate electronic state $6d\ 2D_{3/2}$ (36117.9 cm^{-1}) was performed by a frequency-doubled narrow-band tunable dye laser beam at 276.9 nm with a linewidth of 0.8 GHz. The subsequent ionization of excited thallium atoms was accomplished by a powerful 532.5 nm beam of 10 kHz EdgeWave Nd:YAG laser, that was also used as a pump source for the first excitation step laser (for details of RILIS laser setup see [31]). The non-selective thermal ionization of thallium on a surface of the hot cavity was also present, however it was approximately 20 times less efficient than the laser ionization. The ions were then accelerated to an energy of 30 keV and separated according to their mass to charge ratio by the ISOLDE General Purpose Separator. As a result, a high-purity beam of ^{184}Tl was obtained. Moreover, by tuning the first excitation step frequency of the laser to selectively enhance the production of the (10^-) state, an isomerically purified beam was produced. This was possible due to the marked difference in hyperfine structures of the different ^{184}Tl isomers. After separation, the ion beam entered the Windmill system [34, 35, 36] through a collimator and was then implanted in one out of the ten carbon foils (6 mm diameter, 20 $\mu\text{g}/\text{cm}^2$ thickness [64]) that were mounted on a rotating wheel. After each supercycle the wheel was rotated to move away the radioactivity from the implantation position and present a fresh foil in front of the ion beam. The separator beam gate was opened for different periods during the experimental campaign varying between 14 and 300 ms after the proton pulse in order to limit the amount of activity implanted on the foil.

One Si detector (Si1: total area 450 mm², 300 μm depletion depth) was placed upstream 7 mm away from the implantation foil. The beam passed through a hole of 8 mm diameter. The other Si detector (Si2: total area 300 mm², 500 μm depletion depth) was placed 4 mm behind the implantation foil. These detectors were also used for detecting β^+ particles and conversion electrons (CE). The detection efficiency of Si2 for electrons was determined by using the CE from the 375 keV E0 transition ($0_2^+ \rightarrow 0_1^+$) in ^{184}Hg , where the former is populated in the β^+/EC decay of ^{184}Tl . Due to the limited resolution of the Si2 detector, M(and higher order)-CE could not be resolved from L-CE. This results in 7.6(16)% at 292 keV (K conversion) and 7.2(17)% at 360 keV (L,M conversion) (see [65]). The two Si detectors were placed at the implantation position in close geometry, covering a total solid angle of 24% of 4π . To detect γ

rays two single crystal HPGe detectors were placed outside the vacuum chamber at 0° (Ge1) and 90° (Ge2) respectively with respect to the direction of the incoming beam. The typical energy resolution (full width half maximum) of each crystal for 1.3 MeV γ radiation was ~ 3.1 keV. The absolute photo-peak efficiency for the 506 keV line, the strongest transition in the IT decay of the (10^-) state in ¹⁸⁴Tl, was 4.9(1)% and 0.76(3)% in Ge1 and Ge2 respectively, for the given geometry. Energy and efficiency calibrations were performed using standard sources of ¹³³Ba, ¹³⁷Cs, ⁶⁰Co and ¹⁵²Eu. In the detection setup, digital electronics (digital gamma finder (DGF) modules [67]) were used to acquire the data. Unfortunately, due to the high count rates, we suffered from a dead time effect caused by the buffer read-outs of the DGF modules.

4.1.3 Internal decay of the (10^-) Isomer

During the experimental campaign, three different data sets were acquired. In the first data set, the thallium atoms were ionized only through surface ionization. A second data set consists of laser scans, where decay spectra were taken as a function of the first excitation step laser frequency. And then finally a third data set was acquired, where the first excitation step laser frequency was tuned in order to selectively enhance the production of the (10^-) state and, through its IT, also the (7^+) state relative to the (2^-) state. Combining these different data sets we were able to disentangle the decay schemes of the three long-lived states in ¹⁸⁴Tl [65, 69], namely, the α decay and IT of the (10^-) , the α and EC/ β^+ decay of the (7^+) and the (2^-) .

The presence of different decaying states in ¹⁸⁴Tl can be clearly demonstrated using the intensity as a function of wavenumber for specific γ lines. Fig. 4.1 shows results from the second data set where the intensity evolution of three such γ rays, each of them specific for the decay from one of the three long-lived states, is given as a function of the wavenumber of the first excitation step laserlight. Due to the difference in nuclear properties (spin, magnetic moment, charge radius) of the isomers, the atomic levels are splitting and shifting differently through the hyperfine interaction making the laser-ionization efficiency for the different isomers wavenumber dependent. The 506 keV transition originates purely from the (10^-) state in ¹⁸⁴Tl and leads to a hyperfine structure pattern characteristic for the (10^-) state. The 340 keV γ line is the $6^+ \rightarrow 4^+$ transition in ¹⁸⁴Hg. The former is predominantly fed by the (7^+) β decay in ¹⁸⁴Tl which leads to a hyperfine structure pattern characteristic for the (7^+) state. The 616 keV is the $2_2^+ \rightarrow 2_1^+$ transition in ¹⁸⁴Hg, where the 2_2^+ is predominantly fed by the (2^-) β decay [65]. This leads to a hyperfine structure pattern characteristic for the (2^-) state. In the third data set, the wavenumber (before frequency doubling) of the first step narrowband laser was tuned to 18058.75 cm^{-1} or 18059.05

cm^{-1} , the maxima for the (10^-) state, in order to enhance its production and, through the IT, that of the (7^+) state over the (2^-) state. These wavenumbers are indicated in Fig. 4.1 by the vertical dashed lines.

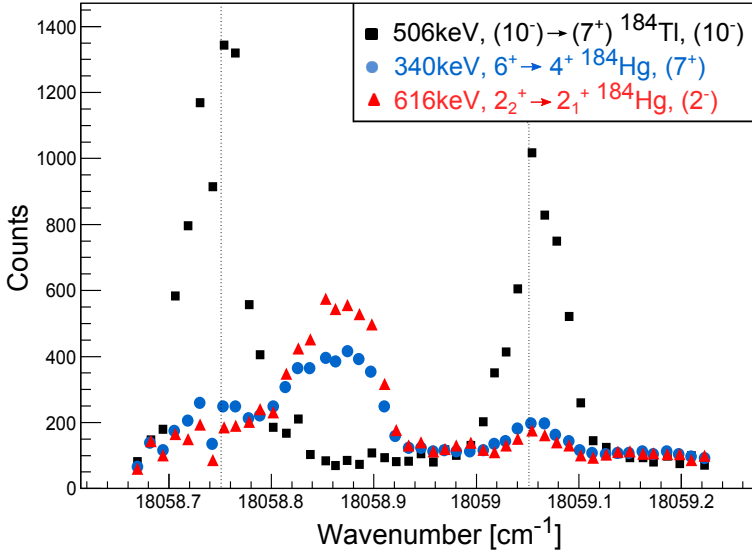


Figure 4.1: [Color] Intensity of the 506 keV (black squares), 340 keV (blue circles) and 616 keV (red triangles) γ lines as a function of the first excitation step laser wave number (before frequency doubling). The origin of the different γ lines is given in the inset and further discussed in the text. The vertical dashed lines indicate the wavenumber (before frequency doubling) to which the first step narrowband laser was tuned to in order to enhance the production of the (10^-) state.

Another way to discriminate between the isomers is by making use of the difference in half-life. This is unfortunately not possible for the (7^+) and (2^-) states as they have similar half-lives ($T_{1/2} \sim 10$ s [76, 77, 78]). However, based on the systematics of the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ to $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ IT in the heavier odd-odd thallium isotopes, a half-life in the order of ~ 100 ms is expected for the (10^-) isomer. The pulsed structure of the proton beam together with the short implantation time after each proton pulse enables the discrimination between short- and long-living activity as is shown in Fig. 4.2. Spectrum 4.2a) is a singles γ -ray energy spectrum collected between 10 and 200 ms after each proton pulse and thus contains relatively more short-living than long-living

activity. Spectrum 4.2b) was acquired starting from 200 ms after each proton pulse until the next proton pulse. Many γ rays are present in both spectra, mostly originating from the β decay of ¹⁸⁴Tl and from longer-lived daughter activities accumulated in the Windmill chamber. Spectrum 4.2c) results from the subtraction of spectra 4.2a) minus 4.2b), after normalization on the 367 keV γ line. The latter is the $2^+ \rightarrow 0^+$ transition in ¹⁸⁴Hg. The resulting spectrum 4.2c) contains only short-living activity. The γ rays indicated with their corresponding energy were attributed to the de-excitation of the (10^-) isomeric state.

Before discussing the (10^-) decay we will address the peaks and dips indicated with the different symbols in Fig. 4.2c). The small peaks at 159 and 198 keV, indicated with a triangle, result from neutron-induced reactions in the surrounding materials. These neutron-induced γ lines have been identified as such since they are already present in the first 5 ms after the proton pulse when the separator beam gate is still closed. Both γ lines are also clearly present in the singles γ spectra of the other thallium isotopes measured during the same experimental campaign. The peak and dips indicated with a full circle originate from a disproportionate subtraction of the long-lived contribution and result in the creation of artificial peaks. However, the origin of the peak at 117 keV, indicated with a square, remains unclear at present.

Using the same procedure used for the singles γ -ray spectra from Fig. 4.2, an electron spectrum detected with the Si2 detector was constructed. This spectrum (see Fig. 4.3, open spectrum) contains the IT electrons associated with the decay of (10^-) state. The peak at 491 keV corresponds to the L,M-CE from the 506 keV transition. The K-CE from the 506 keV and L,M-CE from the 445 keV transitions give rise to the peak around 421 keV, while the 360 keV peak is attributed to the L,M-CE from the 374 keV and K-CE from the 445 keV transitions. The filled spectrum in Fig. 4.3 represents electrons in coincidence with the K_{α_1} line from thallium and is collected between 10 and 200 ms after the proton pulse.

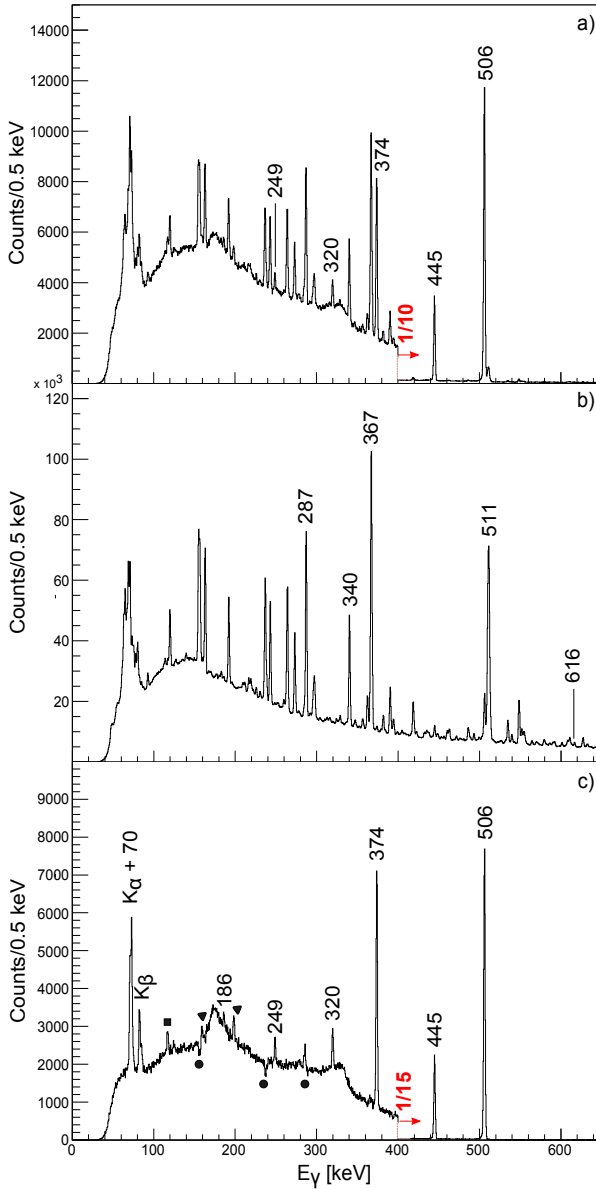


Figure 4.2: [Color] γ -ray singles energy spectra obtained in Ge1 while the first excitation step laser frequency was tuned to enhance the production of the (10^-) state. Spectrum a) is spectrum collected between 10 and 200 ms after each proton pulse, enhancing the IT γ rays (the most intense ones are labeled by their energy). Spectrum b) contains γ rays that were acquired starting from 200 ms after each proton pulse until the next proton pulse, enhancing the γ rays from the β^+/EC decay of ^{184}Tl (labeled by their energy). Spectrum c) is a linear combination of the upper two spectra (see text) and contains dominantly the γ lines associated with the decay of the (10^-) isomeric state. The peaks and dips indicated with different symbols in the spectrum are further discussed in the text.

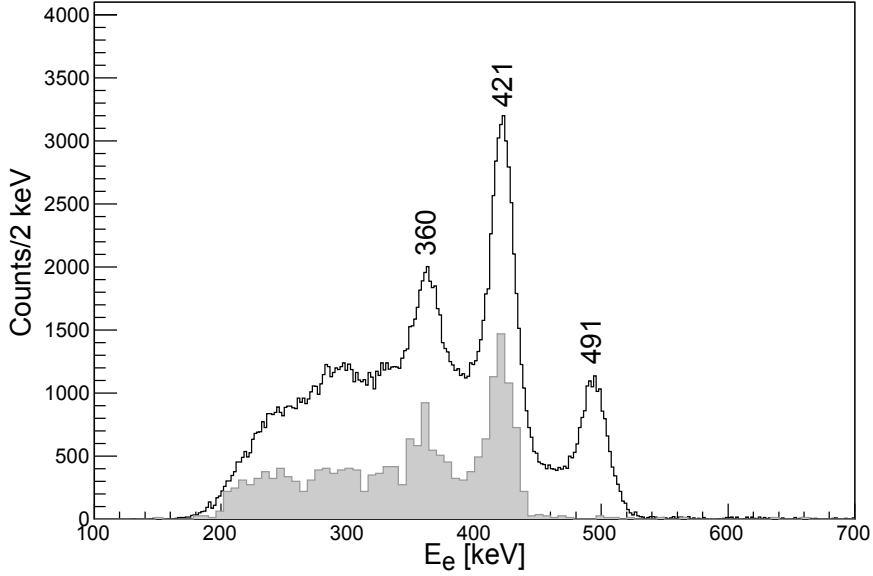


Figure 4.3: Electron spectra containing mainly IT activity. The open spectrum was obtained using the same procedure as used to generate spectrum Fig. 4.2c). The filled spectrum contains only K-CE from the decay of the (10^-) state by applying a coincidence condition on the thallium K_{α_1} X-rays. It is scaled with an arbitrary factor for visualisation purposes. Electron energies are given in keV.

In Fig. 4.4 the time distribution of the events in the 506 keV γ line is shown. The dotted red line shows the result of a fit by an exponential curve with a constant background fixed at 10 counts resulting in a half-life value of 47.6(7) ms. A deviation from the fitted curve is observed for times larger than 250 ms and is due to the previously mentioned dead time effect. Different fits with a constant background varying between 5 and 30 counts were performed. The variation of the resulting half-life values was added as systematic uncertainty to the statistical uncertainty ($\sigma_{stat} = 0.3$ ms, $\sigma_{syst} = 0.4$ ms). The final half-life value of 47.1(7) ms results from the weighted average of the time behavior of the intensity of the γ lines at 506, 445 and 374 keV, the K- and L-CE from the 506 keV transition and the α line from the decay of the (10^-) isomer. The α decay of this isomer, identified for the first time in this study, will be further discussed in [69].

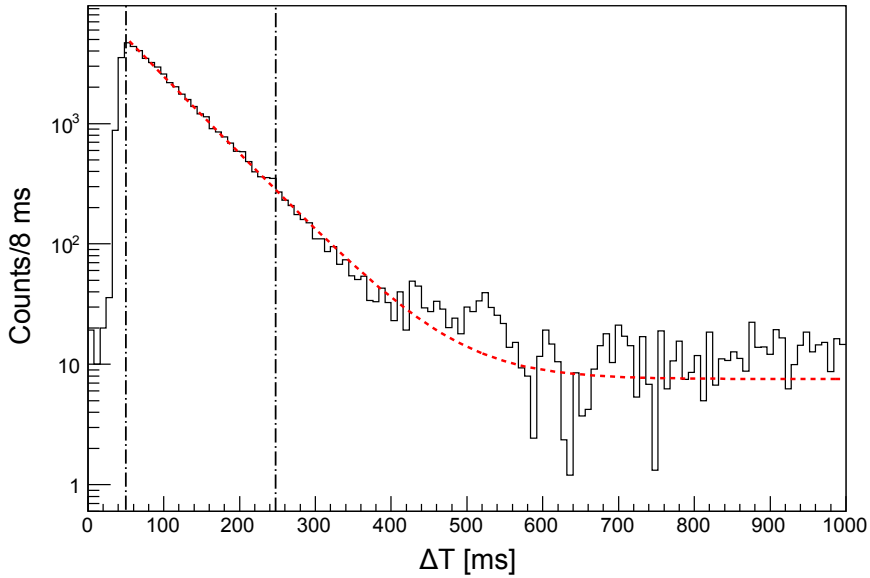


Figure 4.4: [Color] Time distribution of the events in the 506 keV γ line from the (10^-) isomeric state. The dotted red line shows the result of a fit from 50 to 250 ms by an exponential curve with a constant background fixed at 10 counts resulting in the half-life value of 47.6(7) ms. The fit range from 50 to 250 ms is indicated by the vertical dashed lines.

The decay scheme shown in Fig. 4.5 is constructed based on the knowledge of previous experiments [71] and all the γ rays assigned to the IT of the (10^-) state in the singles γ -ray energy spectrum of Fig. 4.2 and Table 4.1, using the Rydberg-Ritz combination principle [79]. Due to low count rates no γ - γ coincidences could be obtained. Spin assignments of the different levels are made based on deduced γ -ray multiplicities. The multiplicities have been attributed by using the γ , electron and K X-ray intensities and the partial half-life of the transitions, starting from the (10^-) IT (see Table 4.1). (Note: From analysis of the α decay of the (10^-) state, it was deduced that the α -branching ratio is negligible when determining the partial half-lives of the γ transitions de-exciting the (10^-) isomeric state [69].) The multiplicity of the 445 keV is deduced by using the intensity of the 360 keV electrons (K-conversion of the 445 keV transition) in the Si2 detector coincident with the thallium K_{α_1} X-rays in Ge1 (see the filled spectrum in Fig. 4.3), the intensity of the 445 keV γ line in singles and the measured efficiencies in the respective Si and Ge detectors. A

K-conversion coefficient of 0.09(2) is obtained: this excludes an E1 multipolarity ($\alpha_{CE,K}(\text{theor.}) = 0.00977(14)$ [55]) but leaves the possibility of $[78(^{+22}_{-26})\% \text{ M1} + 22(^{+26}_{-22})\% \text{ E2}]$ or $[10(^{+16}_{-10})\% \text{ M2} + 90(^{+10}_{-16})\% \text{ E3}]$ [55]. However, out of the intensity ratio between the 360 keV electron line and the 421 keV electron line (K-conversion of the 506 keV transition) in the filled spectrum of Fig. 4.3, only the multipolarity combination E3 for the 506 keV transition ($\alpha_{CE,K}(\text{theor.}) = 0.0489(7)$ [55]) and M1+E2 for the 445 keV transition remains. The E3 character of the 506 keV transition is in agreement with the previously proposed spin and parity of the IT isomeric state at 506 keV (10^-) and the high spin β -decaying state (7^+) [71]. The 445 keV level is fed in the IT of the (10^-) isomer by an unobserved thus highly-converted, parity-changing 61 keV transition. Attributing a pure E3 character to this 61 keV transition would lead to an unphysically low retardation factor F_w of 2.7 (1), while a pure M2 transition leads to a reasonable retardation factor F_w of $3.84 \times 10^4(16)$ (see Table 4.2 for an overview of M2 and E3 retardation factors in the thallium isotopes). Therefore a (8^+) spin and parity is proposed for the level at 445 keV. The level at 320 keV was previously observed by Andreyev et al. [71] through a prompt coincidence between feeding α lines and the γ rays decaying out of this state, restricting the possible multipolarity of the 320 and 249 keV transitions to E1, M1 and E2. Both transitions have the same parity as the 70 keV transition, fed by the 249 keV transition and which is of M1 character [71]. In the IT of the (10^-) isomer, the 320 keV level is fed by the 186 keV transition. Depending on the multipolarity of the 320 and 249 keV transitions, the obtained total conversion coefficient of the 186 keV transition lies between 4.3 and 9.2, allowing M2 and E3 multipolarity but excluding M3. The preference for an E3 multipolarity assignment for the 186 keV transition is based on the associated retardation factor F_w of 486(115) which is consistent with other E3 retardation factors in the thallium isotopes (see Table 4.2), while the retardation factor of $8.5 \times 10^6(20)$ for a M2 transition is rather high. Furthermore, attributing a M2 character to the 186 keV transition would require an unexpected second low-lying 8^+ state. Concluding, a (7^+) spin and parity is preferred for the state at 320 keV. The level at 70 keV was previously observed by Andreyev et al. [71] and a M1 multipolarity was deduced for its de-excitation to the (7^+) β -decaying state. In the (10^-) decay this level is fed through the 249 and 374 keV transitions. The 1% γ -ray intensity of its depopulating 70 keV γ line is in agreement with the previously measured conversion coefficient of 5(1) [71].

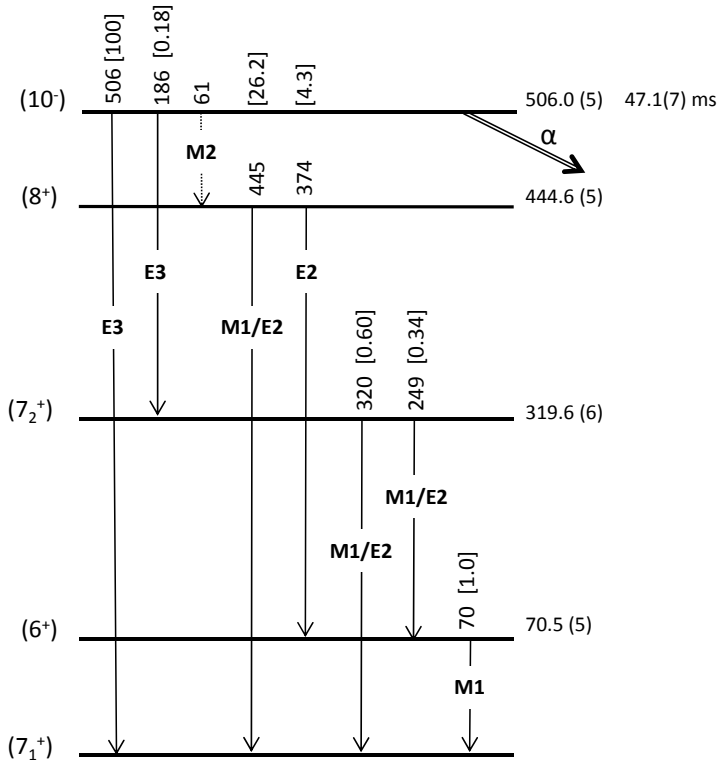


Figure 4.5: Decay scheme of the (10^-) isomer in ^{184}Tl . The energies of the levels and transitions are in keV. The transition intensities (relative to the 506 keV transition intensity) are given in brackets. The deduced multipolarity of each γ line is indicated on the corresponding transition.

4.1.4 Discussion

In the region around neutron mid-shell at $N = 104$ low-lying excited states in the even-mass thallium isotopes result mainly from the coupling of the odd proton occupying the normal $\pi 3s_{1/2}$, $\pi 2d_{3/2}$ or intruder $\pi 1h_{9/2}$ orbitals with the odd neutron in the $\nu 3p_{3/2}$ or $\nu 1i_{13/2}$ orbitals. All the odd-mass thallium isotopes ranging from $A = 201$ down to 181 have a $(1/2^+)$ ground state, a $(3/2^+)$ excited state between 258 and 386 keV and a $(9/2^-)$ intruder state showing a distinct parabolic behavior of the excitation energy as a function of neutron number

Table 4.1: Energy, relative intensity and deduced multipolarity for the γ rays observed in the decay of the (10^-) isomeric state. The relative intensities of $K_{\alpha 1,2}$ and $K_{\beta 1,2}$ X-rays associated with the IT of ¹⁸⁴Tl are also listed.

E_γ (keV)	I_γ (%)	Deduced Multipolarity
506.1 (1)	100	E3
444.8 (1)	26.2 (9)	M1/E2
373.9 (1)	4.3 (1)	E2
319.8 (4)	0.60 (3)	M1/E2
248.9 (2)	0.34 (3)	M1/E2
186.2 (3)	0.18 (4)	E3
70.5 (5)	1.0 (8)	M1
$K_{\alpha 1,2}$	5.9 (4)	
$K_{\beta 1,2}$	1.6 (2)	

reaching its minimum at $N = 108$. The allocation of the states in the odd-odd thallium isotopes within each full multiplet will primarily depend on the specific proton-neutron residual interaction in these $[j_p \otimes j_n] I^\pi$ configurations. This has been thoroughly discussed by several authors [6, 11, 12, 13, 14]. The two lowest states in Fig. 4.5, (7_1^+) and (6^+) , have been proposed in [71] as two members of the $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]_{6^+, 7^+}$ multiplet of states, while the (10^-) is associated with the intruder based $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]_{2^- \rightarrow 11^-}$ configuration. In the present work the levels at 320 [71] and 445 keV have been tentatively assigned spin and parity (7^+) and (8^+) respectively. Both are candidates for members of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]_{5^+ \rightarrow 8^+}$ multiplet. Their energy fits well in the systematics presented in Fig. 4.6, where the position of the $(3/2^+)$ $[\pi 2d_{3/2}]$ level in the neighboring odd-mass thallium isotopes is given. In ¹⁸⁶Tl [71] the levels at 255, 281, 356 and 441 keV were also attributed to the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]_{5^+ \rightarrow 8^+}$ multiplet, although the possibility was left open that the 356 and 441 keV levels are originating as respectively the 5^- and $(6, 7, 8, 9 \text{ or } 11)^-$ members from the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]_{2^- \rightarrow 11^-}$ multiplet.

Next to energy systematics, information on the underlying structure of the different isotopes can be obtained through the reduced widths of the α transitions feeding the levels and through the transition probabilities of the electromagnetic radiation feeding and de-exciting the levels. The α decay of bismuth towards thallium is crossing the $Z = 82$ closed shell and in principle only hindered α decay is expected as the proton configurations are very different. However, allowed α decay is observed and this was used to characterize proton intruder states in the

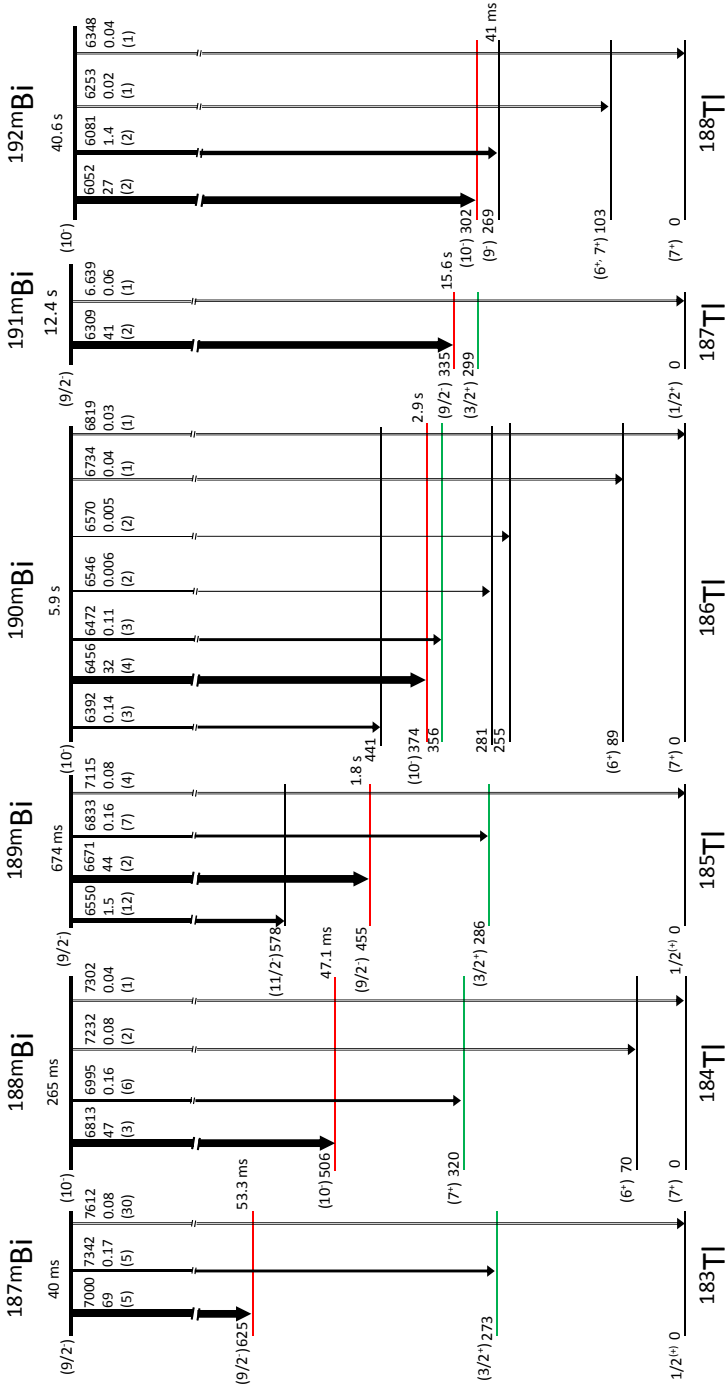


Figure 4.6: [Color] α -decay information from ^{192}Bi down to ^{187}Tl involving the $1h_{9/2}$ proton configuration. The α -decay energy and corresponding reduced width (in keV) are indicated next to each transition. The errors on the reduced widths are shown within brackets. All energies are in keV. The states in the daughter thallium isotopes involving the $\pi 1h_{9/2}$ orbitals are indicated in red, those involving the $\pi 2d_{3/2}$ in green. The data are taken from [13, 14, 15, 71, 80, 81, 82].

bismuth mother and thallium daughter (see [15, 70, 71] and references therein). In Fig. 4.6 the α -decay information from ¹⁹²Bi down to ¹⁸⁷Bi involving the $1h_{9/2}$ proton configuration is presented. This is the decay from the $(9/2^-)$ state for the odd-mass bismuth isotopes and the decay from the (10^-) [$\pi 1h_{9/2} \otimes \nu 1i_{13/2}$] state for the even-mass bismuth isotopes. The reduced widths for α decay have been calculated using the Rasmussen formalism [53]. Five classes of reduced widths can be observed: 20-70 keV, ~ 1.5 keV, ~ 0.16 keV, 0.02-0.08 keV and ~ 0.005 keV. In this mass region, allowed α decay in even-even nuclei typically has reduced widths between 30 and 60 keV. In ^{187–192}Bi, α -decay transitions between two states with equal spin and parity are observed with reduced widths varying between 27 and 69 keV (see Fig. 4.6). The $(9/2^-) \rightarrow (11/2^-)$ decay and $(10^-) \rightarrow (9^-)$ decays have a reduced width around 1.5 keV. The decay towards the $\pi 2d_{3/2}$ state in the odd thallium isotopes has a reduced width around 0.16 keV. In ¹⁸⁸Bi a transition with a similar reduced width feeds respectively a level at 320 keV. In ¹⁹⁰Bi two levels receive similar feeding: the level at 356 keV and at 441 keV. Based on this and their energy position, it is tempting to interpret these levels as originating from the proton $\pi 2d_{3/2}$ configuration coupled to the $1i_{13/2}$ neutron. The decay involving the $\pi 1h_{9/2}$ configuration in the odd-mass bismuth isotopes and the $\pi 3s_{1/2}$ configuration in the thallium daughters is strongly hindered and the reduced widths lie between 0.02 and 0.08 keV. In the even-mass bismuth decay transitions hindered with the same order of magnitude are feeding the lowest (6^+) and (7^+) levels. Based on the same arguments developed above, these levels are interpreted as originating from the $\pi 3s_{1/2}$ proton coupled to the $1i_{13/2}$ neutron. In ¹⁸⁶Tl the levels at 255 and 281 keV are even one order of magnitude less fed and the increased hindrance, around 0.005 keV, is discussed in terms of the angular-momentum coupling between the [$\pi 2d_{3/2} \otimes \nu 1i_{13/2}$] [71]. Unfortunately it was not possible in Ref. [71] to observe similar states in ¹⁸⁴Tl due to the limited statistics. Also in the present study no extra levels around 300 keV were observed possibly owing to the specific decay path of the (10^-) isomeric decay bypassing these levels.

In addition to the α -decay reduced widths the deduced γ -ray transition probabilities between normal and intruder states can give further insight in the underlying nuclear configuration. The retardation factors F_w , the rate of a transition compared to the Weisskopf estimate, of the isomeric E3 and M2 transitions in odd and even mass thallium isotopes are listed in Table 4.2. In the odd-mass thallium isotopes retardation factors for the $(9/2^-)$ to $(3/2^+)$ E3 transition in the order of 100 are observed. This retardation is related to the configuration change of the proton from the $1h_{9/2}$ to the $2d_{3/2}$ orbital ($\Delta\ell = 3$). The retardation of the E3 transition in the odd-odd ¹⁸⁶Tl [12] is however significantly higher. This increase in hindrance factor can be related to the different nature of the stretched E3 transition as it involves configuration

Table 4.2: Energy and deduced multipolarity and corresponding retardation factor F_w for E3 and M2 isomeric transitions in $^{183-188}\text{Tl}$. Data for ^{184}Tl are from this work, the other isotopes from [12, 83].

Isotope	E_γ (keV)	Multipolarity	Retardation factor F_w
^{183}Tl	(352)	E3	(79(1))
^{184}Tl	506	E3	984(38)
^{184}Tl	186	E3	486(115)
^{184}Tl	61	M2	$3.84 \times 10^4(16)$
^{185}Tl	169	E3	119(13)
^{186}Tl	374	E3	6470(447)
^{187}Tl	36	E3	99(2)
^{188}Tl	269	M2	$1.89 \times 10^5(19)$

changes from a $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ to a $[\pi 1s_{1/2} \otimes \nu 1i_{13/2}]$ ($\Delta\ell = 4$). In ^{184}Tl the retardation of the transition from the intruder-based (10^-) state to the (7_1^+) state is about seven times smaller than the equivalent transition in ^{186}Tl . This possibly hints towards increased mixing of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration in the (7_1^+) state. Moreover, the (10^-) $\rightarrow$ (7_2^+) E3 transition being twice as fast as the (10^-) $\rightarrow$ (7_1^+) transition points also to a larger $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ content in the (7_2^+) state compared to that in the (7_1^+). It is interesting to note that also in the hyperfine structure measurements in the same experiment, the g factor of the (7_1^+) state shows increased importance of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration, compared to respective g factors in the heavier odd-odd thallium isotopes [7, 84].

4.1.5 Conclusion

The decay of the (10^-) isomeric state in ^{184}Tl was studied at the ISOLDE facility. The half-life was deduced as $T_{1/2} = 47.1(7)$ ms and its IT decay measured. The precisely determined excitation energy of this state (506.1(1) keV) extends the (10^-) $\rightarrow$ (7^+) energy systematics beyond neutron mid-shell and confirms the parabolic behavior as a function of neutron number. From the half-life value and branching ratios, the retardation factors of the depopulating E3 and M2 transitions were determined and compared with retardation factors in neighboring odd-mass and even-mass thallium isotopes. Combined with the information on the published reduced widths of the α decay of $^{187-192}\text{Bi}$ populating levels in daughter nuclei $^{183-188}\text{Tl}$, a confirmation of the proton

1p-2h intruder character of the (10^-) isomer and an interpretation of the levels in ¹⁸⁴Tl fed in the isomeric decay in terms of $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ and $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ could be obtained.

It is worth to note that the proton intruder interpretation of the (10^-) isomer is supported by the results of the isotopic shift/hyperfine structure measurements in the same experiment [84].

4.1.6 Acknowledgements

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4.2 Influence of dead time on half-life determination of (10^-) state in ^{184}Tl

This section will describe in more detail the problems encountered in the determination of the half-life of the (10^-) state in ^{184}Tl caused by the dead time effect (see section 4.1.3).

Figure 4.7 shows the time distribution, relative to the proton pulse, of the events for γ lines with energies from 1 to 3000 keV. On the x-axis, the time difference between the arrival of a proton pulse and the arrival of a γ ray in the HPGe detector is shown, while the y-axis displays the number of counts. The narrow peak in the first 20 ms is due to signals induced by fast neutrons and does not influence the decay curve. After 20 ms, the growing-in followed by the decay dominated by the 47 ms (10^-) state is clearly visible, however, after 200 ms the decay curve is increasing again where it is expected to have reached a constant decay pattern governed by the ~ 10 s β^+/EC of ^{184}Tl . This unexpected behavior is caused by buffer read-outs of the DGF modules. During the experimental campaign, the data acquisition system was configured to perform a read-out when one of the modules has a full buffer or at the end of a measurement cycle. When the buffers of the DGF modules, which can each store about 1000 events, are emptied, the acquisition of incoming signals is blocked, which may result in a significant dead time. The time distribution of the events in the γ lines decaying from the (10^-) state is influenced by this effect and, as a consequence, the decay curves of these γ lines are not purely exponential. As an example, the time distribution of the events in the 506 keV γ line (events in background left and right subtracted) is shown in Fig. 4.8 where the dotted red line shows the result of a fit by an exponential curve with a constant background. The data clearly deviate from the fit.

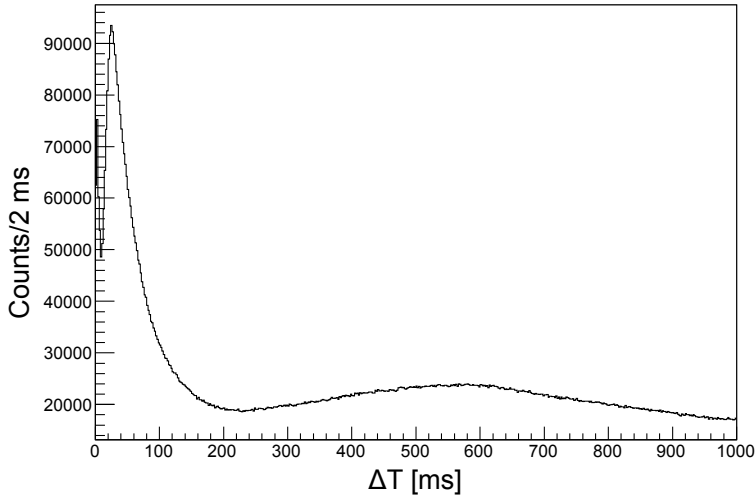


Figure 4.7: Time distribution at $A = 184$ (LI data) after the proton pulse of the events in Ge1 with energies ranging from 1 to 3000 keV.

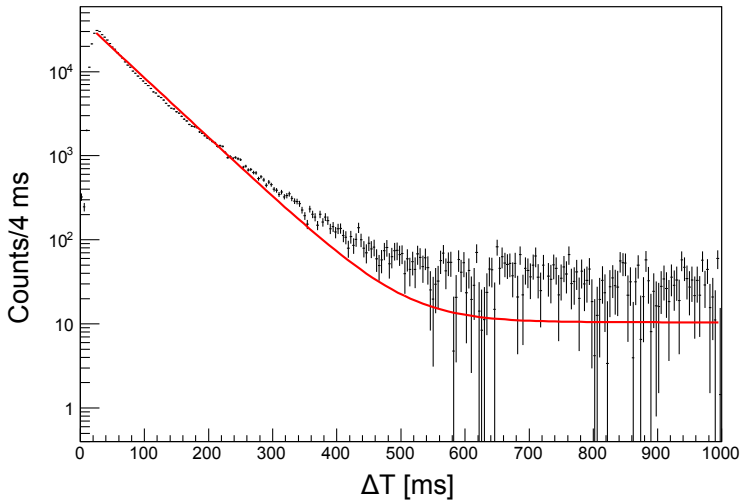


Figure 4.8: [Color] Time distribution of the 506 keV γ -line intensity. The red line shows the result of a fit by an exponential curve with a constant background.

Due to the high count rates after the arrival of a proton pulse, dominated by the production of the (10^-) state in the laser-ionized data set of ^{184}Tl , and the fact that a forced read-out did not take place before the arrival of the proton pulse, the chance for a read-out soon after the arrival of the new proton pulse is high. A read-out takes about 150 ms to complete, which causes a significant amount of crucial data to be lost. As mentioned in section of chapter 3, the dead time effects can be assessed by the use of a 10 Hz pulser to indicate the effective measurement time. This is illustrated in Fig. 4.9 where the black lines represent the arrival of the signal of the 10 Hz pulser and the red lines the arrival of a proton pulse as a function of the start of the data taking. It is clearly seen that, at random moments after the arrival of a proton pulse, the pulser signal is blocked, which points to a read-out of the DGF modules. The top and bottom spectra are for a file in the beginning and at the end of the run respectively, indicating that this problem is persistent throughout the whole run.

This problem can be solved by looking only to the events after the start of a new supercycle since the buffer is emptied at the end of each measurement cycle, instead of including events collected after each proton pulse. This can be seen in Fig. 4.10 where the black lines represent again the arrival of the signal of a 10 Hz pulser and the red lines the arrival of a proton pulse as a function of the start of the data taking. However, in the spectra of Fig. 4.10, the signals at the start of a new super cycle are depicted as opposed to at a randomly selected time during a super cycle in the spectra of Fig. 4.9. The top and bottom spectra are for two different supercycles throughout the measurement file. It should be noted that using the supercycle instead of the proton pulse as a reference timing signal reduces the amount of data significantly, by a factor of about twelve in our case. So in this selection only the most intense transitions can be used to determine the half-life.

In Fig. 4.4 the time distribution of the events in the 506 keV γ line is shown, looking only to the events after the start of a new supercycle. The dotted red line shows the result of a fit by an exponential curve with a constant background fixed at 10 counts resulting in a half-life value of 47.6(7) ms. A deviation from the fitted curve is observed for times larger than 250 ms, indicating buffer read-outs after 250 ms. Thus the fit was limited to the first 250 ms and different fits with a constant background varying between 5 and 30 counts were performed. The variation of the resulting half-life values was added as systematic uncertainty to the statistical uncertainty ($\sigma_{stat} = 0.3$ ms, $\sigma_{syst} = 0.4$ ms). The final half-life value of 47.1(7) ms results from the weighted average of the time behavior of the intensity of the γ lines at 506, 445 and 374 keV, the K- and L-CE from the 506 keV transition and the α line from the decay of the (10^-) isomer.

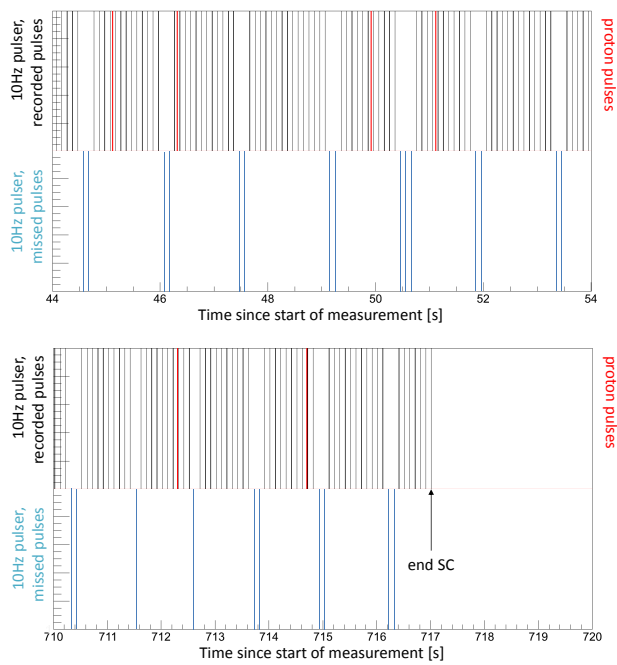


Figure 4.9: [Color] Arrival of a 10 Hz pulser signal (black), proton pulses (red) and the missed pulser signal (blue) as a function of the measurement time. The top and bottom spectra are for a file in the beginning and at the end of the run respectively.

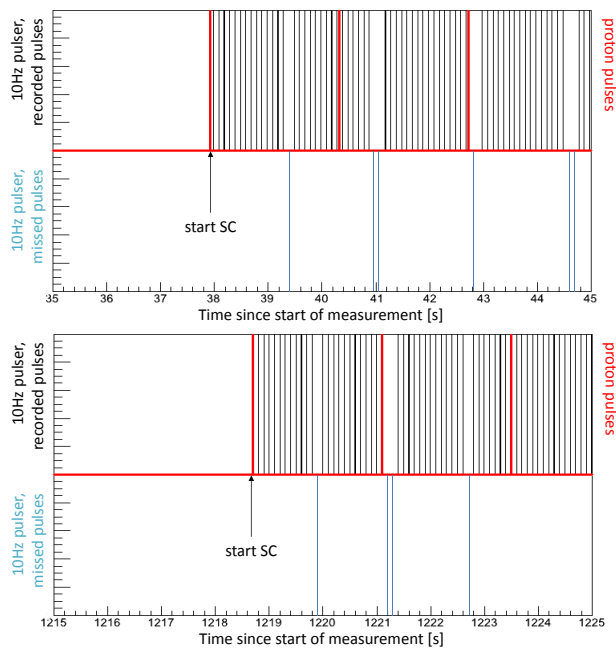


Figure 4.10: [Color] Arrival of a 10 Hz pulser signal (black), proton pulses (red) and the missed pulser signal (blue) at the start of a new SC as a function of the measurement time. The top and bottom spectra are for two different supercycles throughout the measurement file.

4.3 α -decay study of ^{182,184}Tl (Paper II)

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α -decay study of ^{182,184}Tl

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Abstract

α -decay spectroscopy of ^{182,184}Tl has been performed at the CERN Isotope Separator On-Line (ISOLDE) facility. New fine-structure α decays have been observed for both isotopes. α -decay branching ratios of 0.089(19)%, 0.047(6)% and 1.22(30)% have been deduced for the (10⁻), (7⁺) and (2⁻) states respectively in ¹⁸⁴Tl and a lower limit of 0.49% for the α -decay branching ratio of ¹⁸²Tl. A new half-life of 9.5(2) s for the (2⁻) state in ¹⁸⁴Tl and 1.9(1) s for the low-spin state in ¹⁸²Tl has been deduced. Using α - γ coincidence analysis,

multiple γ rays were observed de-exciting levels in $^{178,180}\text{Au}$ fed by $^{182,184}\text{Tl}$ α decays. The γ transitions connecting these low-lying states in $^{178,180}\text{Au}$ are essential to sort the data and possibly identify bands from in-beam studies in these isotopes. Owing to the complex fine-structure α decays and limited knowledge about the structure of the daughter nuclei, only partial level schemes could be constructed for both gold isotopes in the present work. Reduced α -decay widths have been calculated and are compared with values obtained in neighboring odd-A and even-A thallium isotopes. Except for the allowed α decay of the ^{184}Tl (10^-) state, the other fine-structure α decays observed in this study are hindered. This points to strong structural changes between parent thallium and daughter gold isotopes.

4.3.1 Introduction

Although close to the $Z = 82$ shell closure, neutron-deficient nuclei with an odd number of protons and an odd number of neutrons (so called odd-odd nuclei) below lead exhibit complex structures even at low excitation energy. The coupling of an unpaired proton and unpaired neutron gives rise to multiplets of low-lying states from which some can be isomeric [11]. Furthermore this region is notorious for intruder states and shape coexistence [4]. This makes the decay and level schemes of odd-odd nuclei complicated and the spectroscopic interpretation of the underlying configurations of the observed states in many isotopes puzzling. The observation of hindered and unhindered α decay is a powerful method to selectively study and identify these low-lying states in the daughter nuclei, yielding direct information on the excitation energy, decay pattern and possible configurations involved (see e.g. [14, 15, 70, 71] and references therein). In this paper we report on an α -decay study of $^{182,184}\text{Tl}$ ($Z = 81$) which is part of an experimental campaign dedicated to decay and laser spectroscopy studies of neutron-deficient thallium isotopes performed at the Isotope Separator On-Line ISOLDE [33] CERN, Geneva, Switzerland.

Prior to our study, α decay was identified for ^{184}Tl [76] and two α lines of 5988(5) keV and 6162(5) keV were assigned to this isotope based on their similar deduced half-lives of 10(2) s. The authors reported also a third less intense α line around 5815 keV that seemed to decay with a similar half-life. An α -decay branching ratio of 2.1(7)% was extracted using the intensities of both α lines and the intensity of the 367 keV $2^+ \rightarrow 0^+$ transition in ^{184}Hg , which was reported [77] to encompass 80(5)% of the EC/β^+ decay intensity. Alpha decay of ^{184}Tl was also observed by Schrewe et al. [85]. Two α lines with energies 5983(10) and 6155(10) keV were observed, in agreement with those reported by [76], but the authors also identified an α line originating from ^{184}Tl at an energy of 6066(15) keV.

A β -decay study of ¹⁸⁴Tl yielded two β -decaying states, with proposed spin and parity (7^+) and (2^-) based on observed β feeding to the 8^+ as well as to the first excited 2^+ [77]. Both β decays exhibit a half life of 11(1) s [77]. In an α -decay study of ¹⁸⁸Bi [71] three long-living states have been identified in daughter ¹⁸⁴Tl. The (10^-) state in ^{188m1}Bi, of which the assumed major configuration is $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$, decays via an unhindered α decay to a (10^-) state and strongly hindered to a (7^+) state in ¹⁸⁴Tl. The (3^+) $[\pi 1h_{9/2} \otimes \nu 3p_{3/2}]$ state ^{188m2}Bi decays unhindered to a (3^+) state, explained as a $[\pi 1h_{9/2} \otimes \nu 3p_{3/2}]$ intruder state, 99 keV above the weakly fed (2^-) state, explained as regular $[\pi 3s_{1/2} \otimes \nu 3p_{3/2}]$ state. As the relative position of the α -decaying states in ¹⁸⁸Bi is not known [71], the relative position of the (7^+) and (2^-) ¹⁸⁴Tl states are not determined. Direct mass measurements in a Penning trap could not resolve this [72]. In our experimental campaign, an excitation energy of 506.1(1) keV and a half-life of 47.1(7) ms of the intruder-based (10^-) state in ¹⁸⁴Tl have been determined [68]. A review of hindered and unhindered α -decay data of ^{187–192}Bi populating levels in daughter nuclei ^{183–188}Tl including the new information is given in [68] and supports the interpretation of the intruder character of the (10^-) state in ¹⁸⁴Tl.

The first identification of ¹⁸²Tl α decay was by Keller et al. [86] at the velocity filter SHIP. The authors assigned a 6406(10) keV α line to ¹⁸²Tl based on excitation functions. No half-life information was deduced in this study. Bolshakov et al. [78] reported on the observation of 6050 keV α particles from ¹⁸²Tl with a half-life of 2.8(6) s. The authors of [78, 87, 88] deduced an upper limit of 4%, 5% and 5% respectively for the α -branching ratio of ¹⁸²Tl.

Previous α -decay studies of two isomeric states in ¹⁸⁶Bi proposed the existence of two isomeric states in the daughter ¹⁸²Tl [70]. Similar to ¹⁸⁴Tl, the relative position of the α -decaying states in ¹⁸⁶Bi is not known, thus the relative position of the two isomeric states in ¹⁸²Tl is not fixed. Based on a change of decay pattern observed in the neighboring isotope ¹⁸⁸Bi, the authors did not suggest any tentative spin/parity assignments for ¹⁸⁶Bi and ¹⁸²Tl and stress that all possible proton-neutron combinations leading to states with the same spin and parity should be considered.

In the even mass ^{186–204}Tl isotopes, both $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]_{7^+}$ and $[\pi 3s_{1/2} \otimes \nu 3p_{3/2}]$ or $[\pi 3s_{1/2} \otimes \nu 2f_{5/2}]_{2^-}$ configurations have been suggested for the β - and/or α -decaying ground and isomeric states. Only for $A = 190$ and $A \geq 194$ measurements have established $J = 2$ for the ground state spins [89, 90, 91, 92]. For all lighter isotopes, assignments rely on assumptions based on extrapolations using systematics. For the even mass thallium isotopes with $A \geq 186$ $J^\pi = (2^-)$ and $J^\pi = (7^+)$ have been adopted for the ground state and the excited isomeric state respectively and this sequence is extrapolated down to ¹⁸²Tl on systematic grounds [93]. According to [87] a (7^+) assignment for the ¹⁸²Tl

parent state with 3.1(10) s half-life is supported by the observed level population up to 8^+ in ^{182}Hg fed by ^{182}Tl β decay. When moving towards the lighter thallium isotopes with $N = 99$ and lower, a neutron configuration change to $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]_{4^-,5^-}$ is expected. This change was already demonstrated in the neighboring lead, mercury and platinum nuclei at neutron numbers below 100 by, e.g. α -decay studies of $^{179,181}\text{Pb}$ [94, 95, 96] or by observation of low-lying $9/2^-$ (or $7/2^-$ of $\nu 1h_{9/2}$ parentage) states in $N = 99, 101$ mercury and platinum [8]. Therefore a change of the $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]_{7^+}$ to $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]_{4^-,5^-}$ can be expected to occur around $^{180,182}\text{Tl}$. The $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]_{4^-,5^-}$ configuration has been proposed by Elseviers et al. [26] for the ground state of ^{180}Tl in a previous study at ISOLDE, CERN. The presence of the $[\pi 3s_{1/2} \otimes \nu 3p_{3/2}]_{2^-}$ can also not be excluded in ^{182}Tl since it has been established in the heavier odd-odd thallium isotopes. Besides these configurations based on the regular $\pi 3s_{1/2}$ orbital, neutrons in the $i_{13/2}$ and $h_{9/2}$ orbitals can also couple to the intruder $\pi h_{9/2}$ configuration. Similar states have been observed at relatively low excitation energy in odd-odd thallium isotopes with $A \geq 184$ and coexistence of all these, in some cases isomeric, states are thus not excluded in ^{182}Tl . The laser spectroscopy measurements from the present campaign confirm the presence of two long-living states in ^{182}Tl [84] as deduced from differences in the hyperfine structure patterns in both α - and γ -decay data.

Combining the high selectivity of the Resonant Ionization Laser Ion Source (RILIS) [30, 31, 32] with decay spectroscopy and the in-source laser spectroscopy method [73, 74, 75], exotic thallium isotopes down to $N = 97$ have been studied at the ISOLDE facility using the Windmill detection system [34, 35, 36]. Complementary decay data on isomerically purified sources were collected. In the present study, the α -decay characteristics of $^{182,184}\text{Tl}$ are reported. The results on the β decay of $^{182,184}\text{Tl}$ will be reported elsewhere [65].

4.3.2 Experimental setup

The ^{182}Tl and ^{184}Tl α -decay data presented in this study originate from two separate experiments carried out at the CERN Isotope Separator On-Line Device (ISOLDE) facility [33]. The thallium isotopes were predominantly formed through spallation reactions via protons, with an energy of 1.4 GeV and an intensity up to 2.1 μA , bombarding a 50 g/cm² UC_x target. The proton beam consisted of a repeated sequence of pulses (typically 35-40) separated by (sometimes multiple) periods of 1.2 s, referred to as a supercycle. The thallium isotopes were resonantly laser-ionized in the hot-cavity using the same ionization scheme as used in the study of [26, 35]. The excitation from the atomic ground state to an intermediate electronic state $6d\ ^2D_{3/2}$ (36117.9 cm⁻¹) was performed by a frequency-doubled narrow-band tunable dye laser beam at

276.79 nm with a linewidth of 0.8 GHz. The subsequent ionization of excited thallium atoms was accomplished by a powerful 532 nm beam from a 10 kHz repetition rate EdgeWave frequency-doubled Nd:YAG laser, that was also used as a pump source for the first excitation step laser (for details of the RILIS laser setup see [31]). Thermal ionization of thallium in the hot cavity was also present, however it was approximately 20 times less efficient compared to laser ionization. After extraction from the ion source, acceleration to 30 keV and mass separation by the ISOLDE High Resolution Separator (¹⁸²Tl) or General Purpose Separator (¹⁸⁴Tl), a beam of the desired thallium isotopes was transported to the Windmill system [34, 35, 36] through a collimator and was then implanted in one of ten carbon foils (6 mm diameter, 20 $\mu\text{g}/\text{cm}^2$ thickness [64]) mounted on a rotating wheel.

Two silicon (Si) detectors were placed at the implantation position in close geometry, covering a total solid angle of 24% of 4π . An annular silicon detector (Si1: total area 450 mm^2 including a hole of 8 mm diameter, thickness 300 μm) was placed upstream. The beam passed through the hole of 8 mm diameter. This detector was installed for the ¹⁸⁴Tl experiment, but it was absent when ¹⁸²Tl decay data were collected. Another circular silicon detector (Si2: total area 300 mm^2 and thickness 500/300 μm for ¹⁸⁴Tl/¹⁸²Tl measurements) was placed downstream behind the implantation foil. After each supercycle, the irradiated foil was rotated to move away the radioactivity from the implantation position. Longer-lived radiation was detected at the decay position with two circular silicon detectors placed in a similar manner as the implantation detectors (Si3/Si4 total area 300 mm^2 , thickness of 300 μm placed downstream/upstream). In this way measurements at the decay station could be performed while implantation and measurements continued on a fresh foil in front of the ion beam. The energy resolution (full width half maximum, FWHM) of these detectors for α decays in the range of 5000-7000 keV was ~ 25 -30 keV during the ¹⁸⁴Tl experimental campaign and ~ 35 keV during the ¹⁸²Tl campaign. The higher FWHM during the ¹⁸²Tl campaign is due to the fact that this run was dedicated to β -delayed fission (βDF) measurements, which require an energy range of the silicon detectors up to 100 MeV. The absolute efficiency for the Si detectors was determined with two ²⁴¹Am sources mounted on the wheel of the windmill and making use of α - γ coincidences between the 5485 keV ²⁴¹Am α decay and the 60 keV γ ray.

To detect γ rays, two single crystal high-purity germanium (HPGe) detectors were placed outside the vacuum chamber, downstream at 0° (Ge1, 90% relative efficiency, ~ 1 cm from the foil) and at 90° (Ge2, 70% relative efficiency, ~ 3 cm from the foil) with respect to the direction of the incoming beam. The typical energy resolution (FWHM) of each crystal for 1.3 MeV γ radiation was ~ 3.1 keV. Energy and efficiency calibrations were performed using standard calibrated

sources of ^{133}Ba , ^{137}Cs , ^{60}Co , ^{241}Am and ^{152}Eu . The absolute photo-peak efficiency for the 506 keV line, the strongest transition in the internal transition (IT) decay of the (10^-) state in ^{184}Tl , was 4.9(1)% and 0.76(3)% in Ge1 and Ge2 respectively, for the given geometry. Due to the combination of a high γ -multiplicity in the β decay of ^{182}Tl and ^{184}Tl and close-detector configuration, the effective efficiency for the HPGe detectors was lower than that determined by the efficiency calibration using standard calibrated sources. To account for this effect, the efficiency for the 351 and 367 keV γ lines, the $2^+ \rightarrow 0^+$ transition in the β -decay daughters ^{182}Hg and ^{184}Hg respectively, has been determined by γ - γ coincidences. With this method, an efficiency for the 351 keV line in ^{182}Hg of 0.77(7)% and 0.44(4)% was obtained for Ge1 and Ge2 respectively. For the 367 keV line in ^{184}Hg , efficiencies of 2.0(2)% and 0.71(7)% were obtained for Ge1 and Ge2 respectively. The difference in effective efficiency is due to the higher multiplicity in the β decay of ^{182}Tl compared to ^{184}Tl .

In the detection setup, digital electronics (digital gamma finder (DGF) modules[67]) were used to acquire the data[35]. The data were collected event by event and independently time stamped. The event coincidences and time structure were reconstructed by software analysis.

4.3.3 Results

^{184}Tl

During the experimental campaign, three different data sets were collected on mass 184. In the first data set, the thallium atoms were ionized only through surface ionization. This data set is denoted further as Surface Ionized (SI) data. A second data set consists of laser scans, where decay spectra were taken as a function of the first excitation step laser frequency. A third data set was acquired where the first excitation step laser frequency is tuned in order to selectively enhance the production of the (10^-) state and, through its IT, also the (7^+) state relative to the (2^-) state. This data set is denoted further as Laser Ionized (LI) data. In the SI α - and β -decay data set, the ^{184}Tl beam is composed of 61(1)% (2^-) and 39(1)% (7^+) states, while in the LI α - and β -decay data set this is 20(1)% (2^-) and 80(1)% (7^+) states respectively. For a more detailed description of this LI data set we refer the reader to [68]. Combining the different data sets enables to disentangle the decay schemes of the three long-lived states in ^{184}Tl [65, 68], namely, the α decay and IT of the (10^-) , the α and EC/ β^+ decay of the (7^+) and the (2^-) .

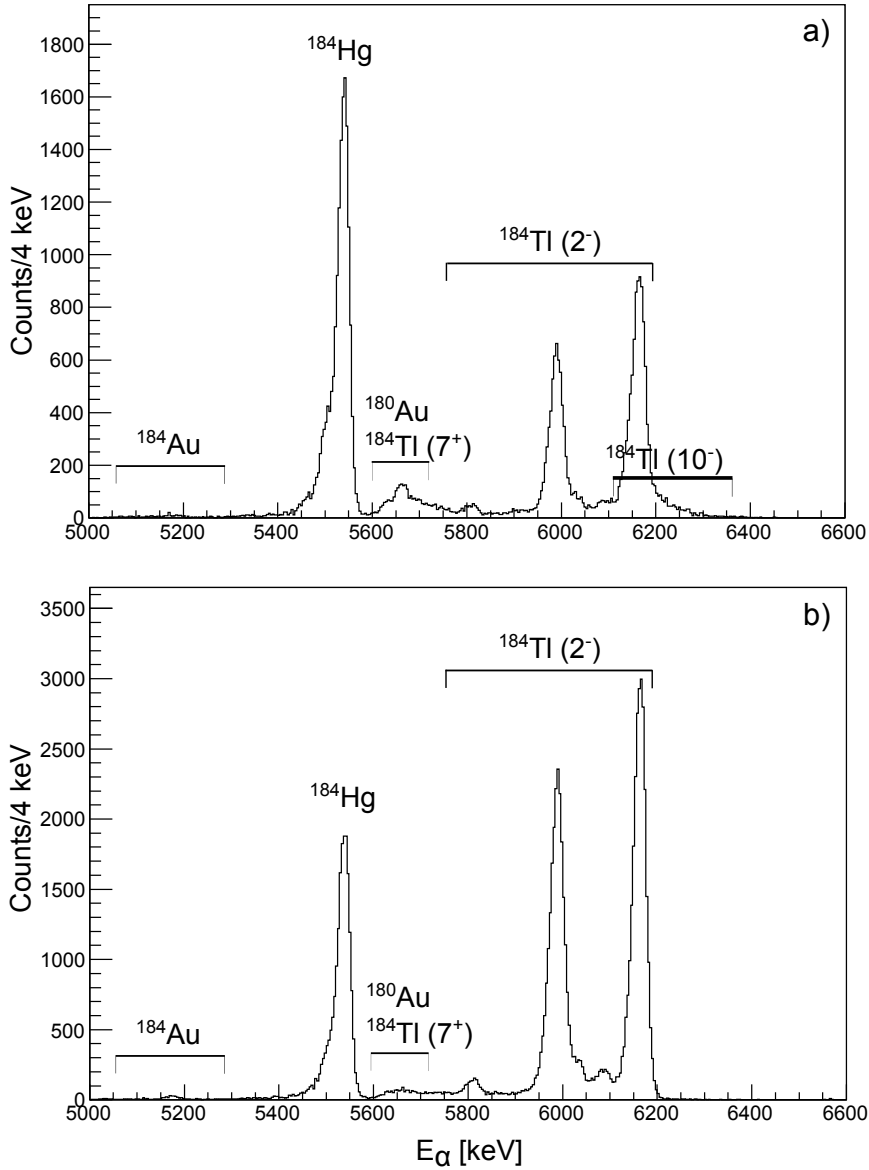


Figure 4.11: Relevant part of the singles α -decay energy spectrum collected at the implantation position (sum of Si1 and Si2). Spectra a) and b) correspond to the LI and SI data sets respectively. The α peaks are denoted with the isotope (and possible spin) to which they belong. The peaks at 5539(5), 5109(5), 5187(5), 5648(10) keV originate from the known α decays of ^{184}Hg , ^{184}Au , ^{184}Au , ^{180}Au respectively [97, 98, 99].

Figure 4.11 shows the singles α -decay energy spectra for mass 184 collected at the implantation position. Spectra 4.11a) and 4.11b) correspond to the LI and SI data sets respectively. The strongest peaks in both spectra are due to known α decays of the isotopes ^{184}Tl and ^{184}Hg , the latter being populated in the β decay of ^{184}Tl . The 5539(5) keV α line from ^{184}Hg and 5987(4) and 6161(4) keV α lines from ^{184}Tl are used for energy calibration [8]. It should be noted however that the α -decay peaks originating from ^{184}Tl are broadened compared to the peak from ^{184}Hg due to α -electron (α -e $^-$) summing effects, where the energy of the α decay, feeding an excited state, which decays by an (at least, partially) converted γ transition, may sum up with the energy of the subsequent conversion electron if both are registered by the same detector. Such summing effects have also been discussed in [81, 100, 101] and we refer to these papers for more details. This summing effect makes the determination of the α -decay energy for the affected peaks difficult and large systematic uncertainties are necessarily introduced on these energy values. Some weaker peaks originating from the α decay of $^{180,184}\text{Au}$ are also identified. The nucleus ^{184}Au is populated in the β decay of ^{184}Hg , while ^{180}Au is, as will be shown later, mainly populated through the α decay of the (2^-) state of ^{184}Tl . A marked difference between both spectra is visible in the extra tail around 6200 keV and the increased intensity around 5650 keV in the LI data, spectrum 4.11a). In the following sections we will focus on the α decay of the three long-lived states in ^{184}Tl .

α decay of the (10^-) isomeric state In the LI data set, the production of the (10^-) and (7^+) states is selectively enhanced relative to the production of the (2^-) state. From the time distribution of the events in the IT of the (10^-) state a half-life of 47.1(7) ms was extracted in our complementary study [68], while the half-lives of the (7^+) and (2^-) states are in the order of ~ 10 s [76, 77, 78]. Using this difference in half-life between the (10^-) and $(7^+, 2^-)$ states within the LI data set, an α -decay energy spectrum containing only α decays associated with the decay of the (10^-) isomeric state can be produced. The pulsed structure of the proton beam together with the short implantation time after each proton pulse enables this discrimination. Figure 4.12 shows a purified (10^-) singles α -decay energy spectrum obtained by subtracting from the singles α -decay energy spectrum collected between 10 and 200 ms after each proton pulse, the singles α -decay energy spectrum collected after 200 ms. Normalization is performed by matching the intensity of the 367 keV γ line in the singles γ -ray energy spectra obtained under the same conditions (see [68]). The 367 keV γ line is the $2^+ \rightarrow 0^+$ transition in ^{184}Hg observed in the β decay of the (7^+) and (2^-) states. In the singles α -decay energy spectrum of Fig. 4.12 an α line at 6132 keV originating from the (10^-) isomeric state is identified. The high-energy tail originates from α -e $^-$ summing. From the time distribution of the events in the α peak, a half-life of 46.5(52) ms is extracted which is consistent with the

half-life of 47.1(7) deduced from the IT of the (10^-) state [68]. The purity of the spectrum of Fig. 4.12 allows the determination of the α -decay branching ratio for this isomeric state. By comparing the intensity of the 6132 keV α decay and the 506 keV γ ray of the (10^-) state, an α -decay branching ratio of 0.089(19)% is deduced, using an absolute γ -decay branching ratio of 70(3)% of the 506 keV γ transition [68]. Using the Rasmussen formalism [53] this leads to a reduced α -decay width of 16(4) keV, indicating an unhindered α decay to a state in daughter ¹⁸⁰Au.

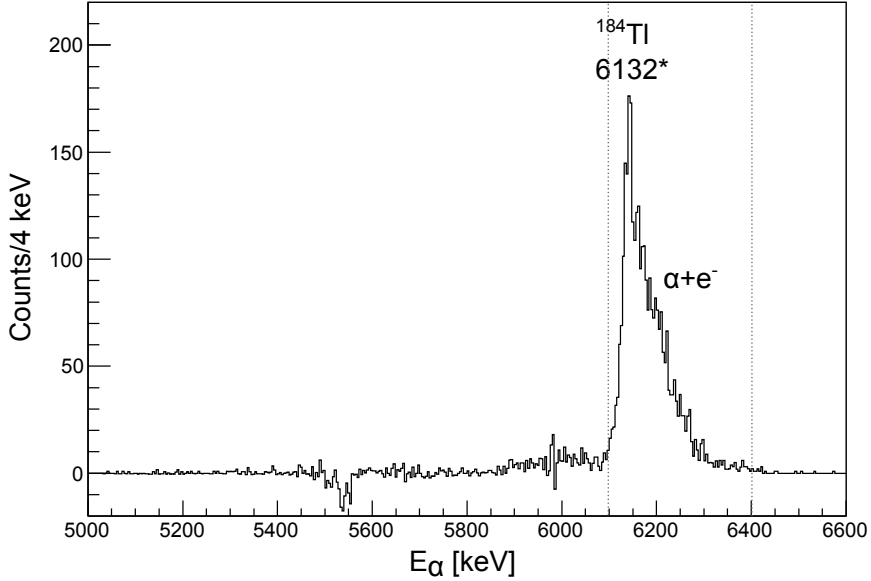


Figure 4.12: Relevant part of the purified α -decay energy spectrum from the (10^-) isomeric state in ¹⁸⁴Tl collected in Si1 and Si2. The α -decay energy is denoted in keV. The vertical dashed lines indicate the gate used for α - γ coincidences (see Fig. 4.13). (*: energy determined from α - γ coincidences)

Figures 4.13a) and b) show the α - γ coincidence matrix and the projection on the γ -energy axis focussing on the (10^-) isomeric state, using a prompt coincidence window of 500 ns. Both spectra are produced using the LI data set and selecting events collected between 10 and 200 ms after each proton pulse. For the (10^-) state, one can distinguish five groups of α - γ coincident events. The α line at 6132 keV is seen in coincidence with four main γ lines at 101, 108, 163 and 206 keV and Au K X-rays. Comparison of the peak intensity ratio in the Au K

X-ray region with the theoretical K_α/K_β ratio reveals a γ transition of about 78 keV buried under the Au K_β X-rays. At the right-hand side of the α peak in the singles spectrum of Fig. 4.12 a tail is present. This tail originates from the previously mentioned α - e^- summing effects. The 78, 101, 108 and 163 keV γ rays are seen in prompt coincidence with these α - e^- summed events, which indicates that these transitions decay most probably as part of different cascades. However, the 206 keV γ line is only coincident with the low-energy part of the α peak and not with the α - e^- tail. This observation suggests that the 206 keV γ line probably feeds a long-lived state in daughter ^{180}Au . No de-exciting γ rays of this state were seen in coincidence with the (10^-) α line using a (delayed) coincidence gate up to 1 ms. The prompt coincidence condition limits the possible multipolarity of all five γ rays to E1, M1 or E2. An overview of the observed α decays and coincident γ rays for the (10^-) isomeric state is given further in Table 4.3. Due to the limited statistics and lack of knowledge about the structure of daughter isotope ^{180}Au , only a partial α -decay scheme can be constructed for this isomeric state (see discussion).

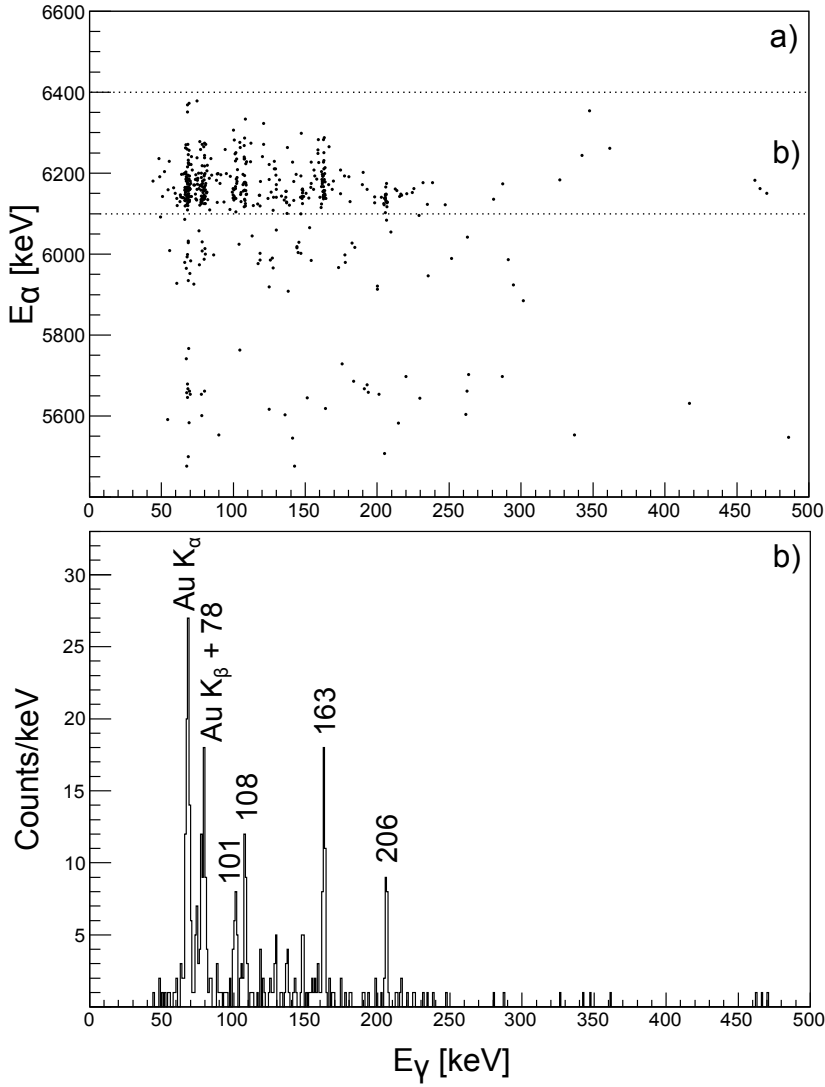


Figure 4.13: a) the prompt α - γ coincidence matrix at mass 184, collected between 10 and 200 ms after each proton pulse, using the LI data set. b) the projection on the γ -energy axis for the events between the horizontal dashed lines corresponding to the (10^-) state (see also Fig. 4.12). The energy of the coincident γ rays is indicated in keV.

α decay of the (7^+) isomeric state To discriminate between the decay of the (7^+) and (2^-) states, one can combine the SI and LI data sets. Within both data sets only events are selected that are collected from 200 ms after the proton pulse until the next proton pulse to eliminate the activity from the short-living (10^-) state. As mentioned in the previous section, in the LI data set the activity from the (7^+) is enhanced over that of the (2^-) state compared to the SI data. The purified (7^+) α -decay energy spectrum in Fig. 4.14 results from the subtraction of the singles α -decay energy spectrum in the SI data from the singles α -decay energy spectrum in the LI data, after normalization using the 6161 keV α -decay intensity that originates from the α decay of the (2^-) state in ^{184}Tl . Using the same procedure, a purified (7^+) γ -ray spectrum can be obtained (see also [65]). In Fig. 4.14 an α line at 5659 keV, together with an α -e $^-$ tail, originating from the (7^+) isomeric state in ^{184}Tl is identified. The peak at 5539 keV originates from the α decay of ^{184}Hg , now exclusively fed through the β decay of the (7^+) state in ^{184}Tl . The half-life of the (7^+) isomeric state can not be determined due to the presence of an α line from the α -decaying daughter ^{180}Au that has a similar α -decay energy of 5648 keV and 8.1(3) s half-life [99] in the singles α -decay energy spectrum (see Fig. 4.11). To calculate the reduced α -decay width the literature value of 11(1) s [77] is used for the half-life. An α -decay branching ratio of the (7^+) state of 0.047(6)% is deduced by comparing the α -decay intensity in the purified (7^+) α -decay energy spectrum with the intensity of the 367 keV $2^+ \rightarrow 0^+$ transition in ^{184}Hg in the purified (7^+) γ -ray energy spectrum. An absolute β -decay branching ratio of 90(2)% of this transition in the (7^+) β decay, as reported in [65], is used. Using the Rasmussen formalism [53], a reduced α -decay width of 4.9(12) keV is determined, indicating a hindered character of the 5659 keV α decay to a level in ^{180}Au .

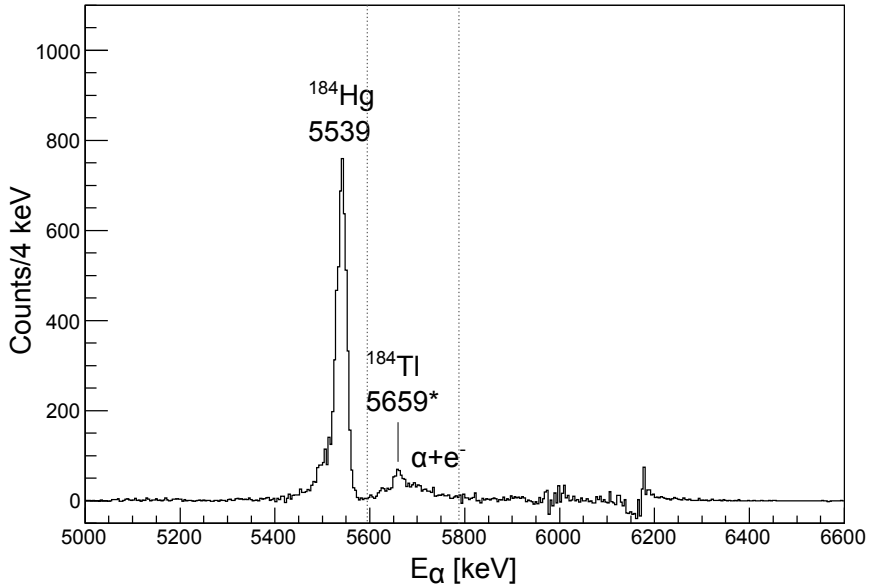


Figure 4.14: Relevant part of the purified α -decay energy spectrum from the (7^+) isomeric state in ^{184}Tl , collected in Si1 and Si2. The α -decay energies are denoted in keV. The vertical dashed lines indicate the gate used for α - γ coincidences (see Fig. 4.15). (*: energy determined from α - γ coincidences)

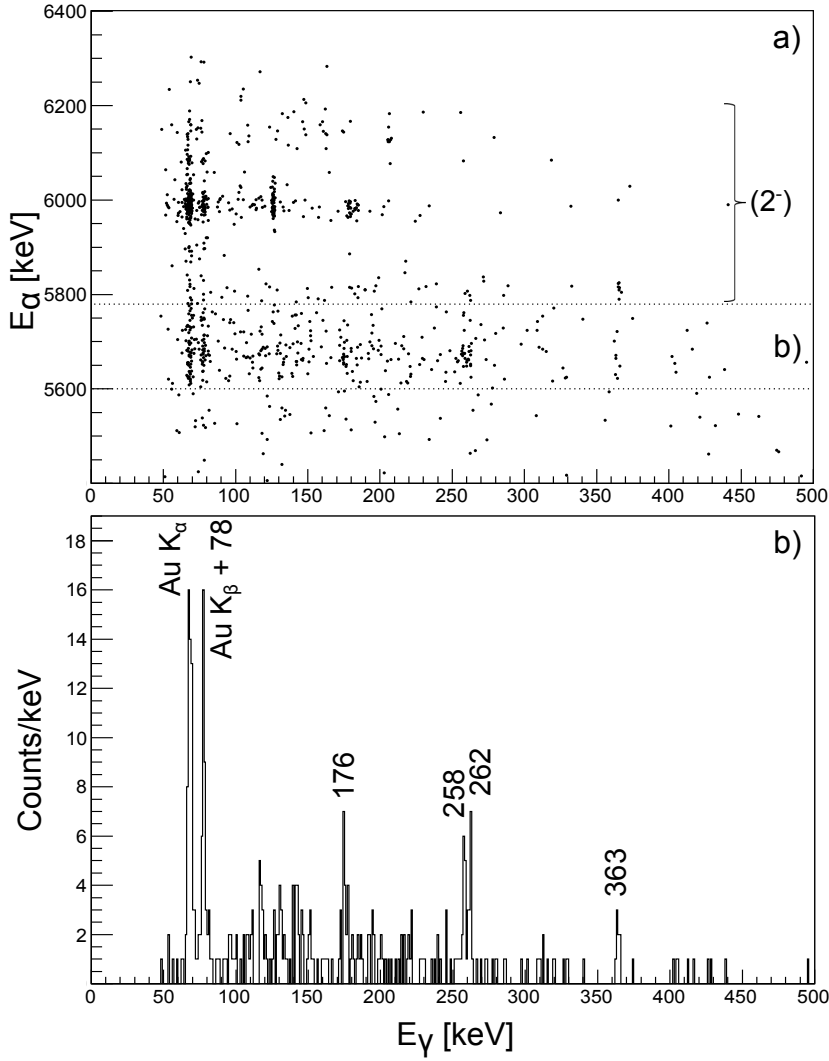


Figure 4.15: a) the prompt α - γ coincidence matrix at mass 184, using the LI data and selecting events collected from 200 ms after the proton pulse. b) the projection on the γ -energy axis for the events between the horizontal dashed lines defining the (7^+) events (see also Fig. 4.14). The energy of the coincident γ rays is indicated in keV.

In Fig. 4.15 the α - γ coincidence matrix focussed on the (7^+) isomeric state is shown in 4.15a) and the corresponding projection on the γ -energy axis in 4.15b). A prompt coincidence window of 500 ns is used. Both spectra are produced using the LI data set and selecting events collected from 200 ms after the proton pulse on until the next proton pulse. It should be noted that some events originating from the (2^-) α decay are also observed, due to surface ionization which is always present (see earlier). Five groups of α - γ coincident events can be identified. The γ lines at 363, 262, 258, 176 and Au K X-rays are seen in coincidence with the α line at 5659 keV. Also in this case a γ transition of 78 keV buried under the Au K $_{\beta}$ X-rays is revealed when comparing the peak intensity ratio in the Au K X-ray region with the theoretical K_{α}/K_{β} ratio. Due to the limited statistics and lack of knowledge about the structure of daughter isotope ¹⁸⁰Au, only a partial α -decay scheme can be constructed for this isomeric state (see discussion). An overview of the observed α decays and coincident γ rays for the (7^+) state is given in Table 4.3.

α decay of the (2^-) isomeric state Using a similar procedure as in the case of the (7^+) isomeric state, a singles α -decay energy spectrum containing only α decays associated with the decay of the (2^-) state can be produced. The spectrum in Fig. 4.16 results from the subtraction of the singles α -decay energy spectrum in the LI data from the singles α -decay energy spectrum in the SI data, after normalization on the 340 keV γ line. The latter is the $6^+ \rightarrow 4^+$ transition in ¹⁸⁴Hg and is predominantly fed by the (7^+) β decay in ¹⁸⁴Tl. Also for the (2^-) state, singles γ -ray energy spectra are produced using the same purification procedure. It should be noted that a normalization factor using the α intensity of the (7^+) peak cannot be used due to the presence of the ¹⁸⁰Au α line and possibly α intensity from the (2^-) decay buried under that of the ¹⁸⁴Tl (7^+) (see Fig. 4.11). The peak at 5539 keV originates from the α decay of ¹⁸⁴Hg, fed by the β decay of the (2^-) state in ¹⁸⁴Tl. The previously suggested [76] α lines decaying from the (2^-) state in ¹⁸⁴Tl, seen in our work at 5810, 5988, 6161 keV are indicated in the singles spectrum from Fig. 4.16.

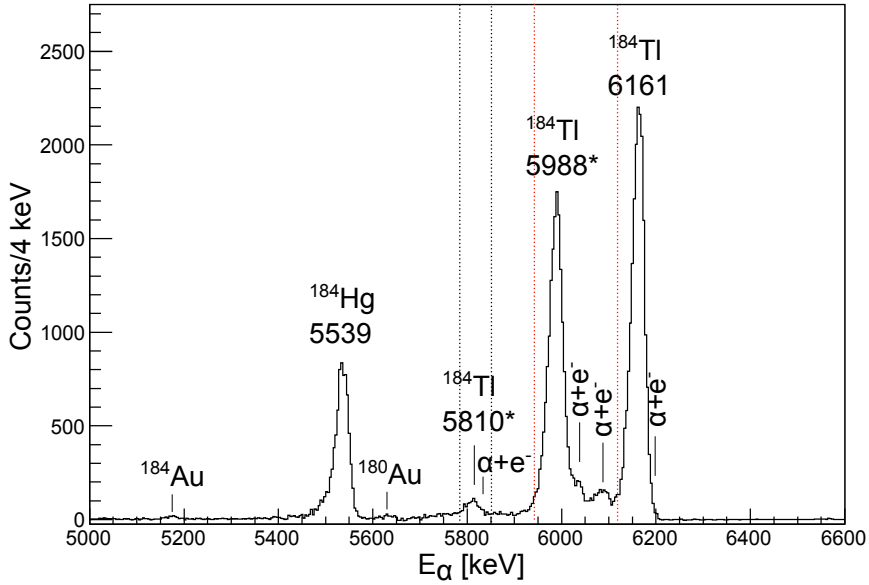


Figure 4.16: [Color] Relevant part of the purified α -decay energy spectrum from the (2^-) isomeric state in ^{184}Tl , collected in Si1 and Si2. The α -decay energies are denoted in keV. The vertical dashed lines indicate the gates used in α - γ coincidence analysis (see Fig. 4.18). (*: energy determined from α - γ coincidences)

Figures 4.17a) and b) show the time distribution of the events in the 5950-6010 and 6140-6200 keV α lines from the (2^-) isomeric state respectively. Data are collected at the decay position (Si3 and Si4) and are not affected by the incoming beam. The dotted red line results from a fit of a simple exponential function to the data resulting in half-life values of 9.4(2) s and 9.6(2) s for a) and b) respectively, which leads to a mean value of 9.5(2) s.

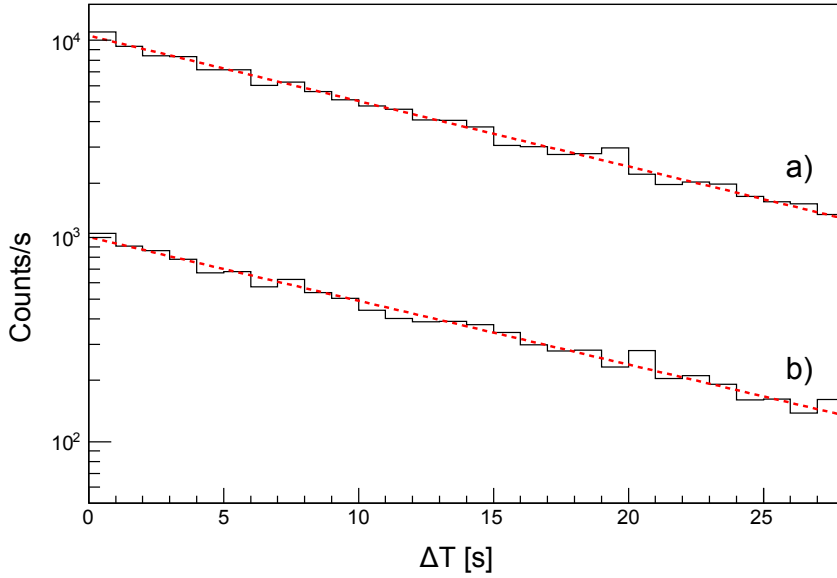


Figure 4.17: [Color] a) and b) show the time distribution of the events in the 5950-6010 and 6140-6200 keV α lines from the (2^-) isomeric state respectively, collected at the decay position (Si3 and Si4). The data indicated with a) are upscaled by a factor of ten for visualization purposes. The dotted red lines result from a fit of a simple exponential function to the data.

The α -decay branching ratio of the (2^-) state is deduced by comparing the intensity ratio of the total number of α decays attributed to ^{184}Tl (7^+) or (2^-) relative to the number of α decays from ^{184}Hg in respectively the purified (7^+) and (2^-) spectra. By using the measured α -decay branching ratio of the (7^+) state (0.047(6)%), an α -to- β normalization factor can be deduced between the number of α particles from the (7^+) decay and from ^{184}Hg (β daughter of ^{184}Tl). As the (2^-) and (7^+) half-lives are similar, the same normalization can be used to deduce the α -branching ratio of the (2^-) decay, resulting in a value of 1.22(30)%. This value includes a systematic uncertainty of 20% to take into account a possible difference in half-life between the (2^-) and (7^+) states. Using the Rasmussen formalism [53] reduced α -decay widths are calculated for the different decay branches. These results will be discussed further.

More α lines are identified by detailed α - γ coincidence analysis (see Table 4.3). Figure 4.18 shows the prompt α - γ coincidence matrix focussed on the (2^-) isomeric state and two projections on the γ -energy axis corresponding to the

two regions between the horizontal dashed lines in the α - γ coincidence matrix. All spectra are produced using the SI data set, an α - γ prompt window of 500 ns and selecting events collected from 200 ms after the proton pulse on until the next proton pulse. No coincidence relation between the α line at 6161 keV and any γ rays is observed. An overview of the observed α decays and coincident γ rays for the (2^-) state is given in Table 4.3. Owing to the complexity of the α -decay pattern and limited knowledge on the structure of daughter isotope ^{180}Au , only a partial α -decay scheme can be constructed (see discussion).

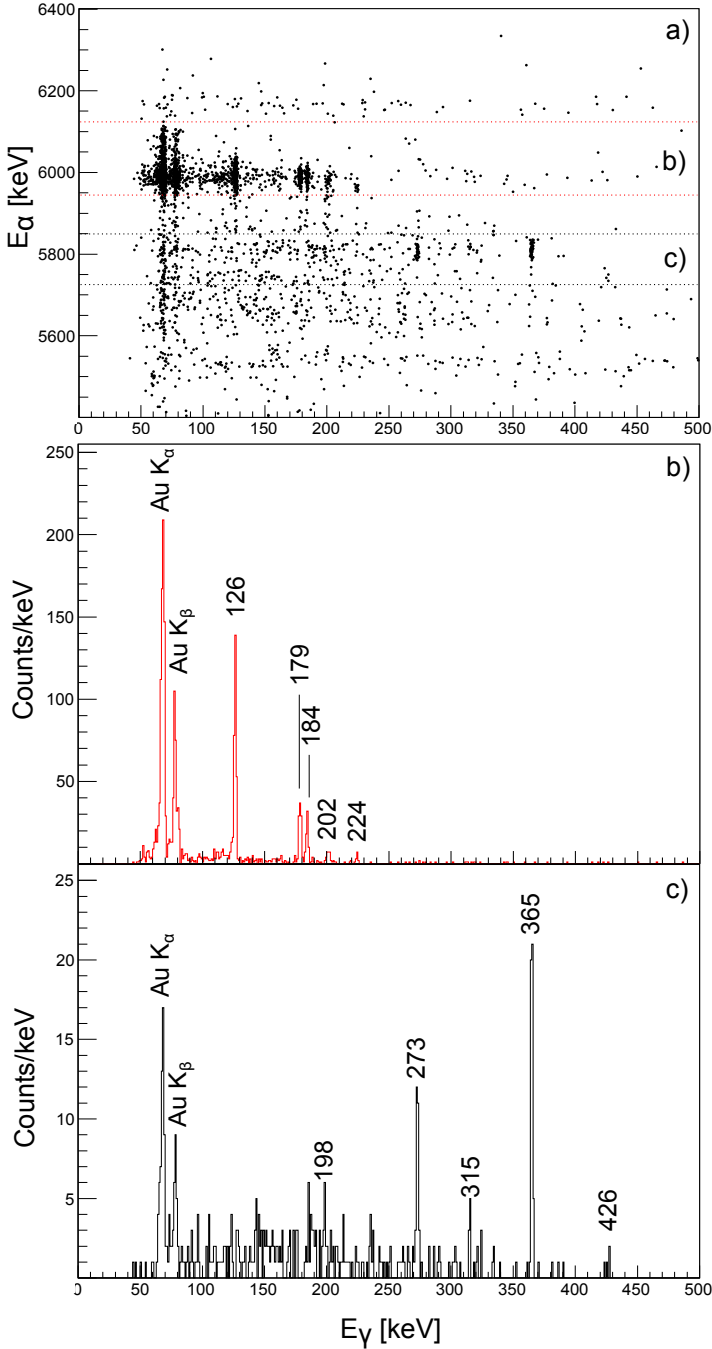


Figure 4.18: [Color] a) the prompt α - γ coincidence matrix at mass 184, using the SI data and selecting events collected from 200 ms after the proton pulse. b) and c) projections on the γ -energy axis for the events between the horizontal dashed lines defining the (2^-) events (see also Fig. 4.16). The energy of the coincident γ rays is indicated in keV.

¹⁸²Tl

Figure 4.19 shows the relevant part of the singles α -decay energy spectrum taken at mass 182. Five α lines are assigned to the decay of ¹⁸²Tl (see further) and are indicated in Fig. 4.19. Only the ¹⁸²Tl peaks observed in our work at 6046 and 6406 keV are known from literature [78, 86]. The other peaks in Fig. 4.19 originate from the α decay of ¹⁸²Hg, ¹⁷⁸Pt and ¹⁸²Au, all expected decay products of ¹⁸²Tl. The 5446(3), 5867(5) and 6406(10) keV α lines from ¹⁷⁸Pt, ¹⁸²Hg and ¹⁸²Tl, respectively, are used for calibration [8]. From the time distribution of the events in the peaks at 6360 keV and 6406 keV, a half-life of 1.9(1) s has been extracted (see Fig. 4.20). This is noticeably smaller and more precise than the previously reported values of 2.8(6) s [78] and 3.1(10) s [87] from α - and β -decay data respectively, but agrees with the value of 2.0(3) s reported by [78] deduced from a β -decay study. The half-lives determined from the other three peaks are consistent with the value of 1.9(1) s, supporting their assignment to ¹⁸²Tl.

In accordance with the results of the α -decay study of ¹⁸⁶Bi [70], clear evidence was found for the presence of two isomeric states in ¹⁸²Tl from β -decay studies [65]. The hyperfine structure patterns for different α - and γ -lines in the ¹⁸²Tl decay spectra, obtained in narrow-band laser scans, strongly support this observation [84]. The hyperfine structure pattern of the $8^+ \rightarrow 6^+$ (413 keV) and $6^+ \rightarrow 4^+$ (332 keV) γ transitions in β -decay daughter ¹⁸²Hg, as a function of laser frequency is distinctly different from the pattern of the $2^+ \rightarrow 0^+$ (351 keV) transition, indicating a high-spin β -decaying state and a low-spin β -decaying state. All α lines in our study have a hyperfine structure pattern testifying to the predominance of the low-spin state decay in these lines. Out of the narrow-band laser scans, there is evidence for α decay from the high-spin state in the region around 6250 keV, however no further information can be extracted. We refer the reader to [84] for a more detailed description of the hyperfine structure of these two decaying states in ¹⁸²Tl. In the complementary β -decay study of the same data set [65], it was not possible to disentangle the two β -decay schemes. As a consequence, we are only able to deduce a lower limit for the α -branching ratio of the low-spin state in ¹⁸²Tl. This lower limit of 0.49% is deduced by comparing the ¹⁸²Tl intensity in the singles α spectrum with the intensity of the 351 keV $2^+ \rightarrow 0^+$ transition in ¹⁸²Hg in the singles γ spectrum, using the absolute β -branching ratio of 74(3)% of this transition in the β decay from [65]. This lower limit for the branching ratio is the edge within which 95% of the events will be found.

During the experimental campaign, one of the goals was to search for the β DF decay of ¹⁸²Tl. However, no fission events are observed in this data set. Considering that less than one β DF fission event is observed, a lower limit for

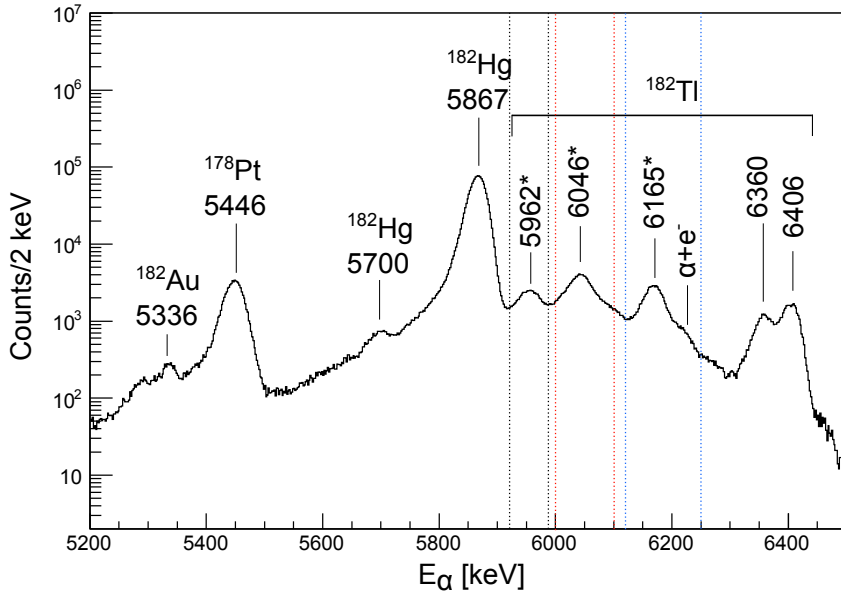


Figure 4.19: [Color] Relevant part of the α -decay energy spectrum at the implantation position measured at mass 182 (Si2). The α -decay energies are denoted in keV. The vertical dashed lines indicate the gates used in α - γ coincidence analysis (see Fig. 4.21). The peaks at 5700(8), 5867(5), 5336(8) and 5446(6) keV originate from the known α decay of ^{182}Hg , ^{182}Hg , ^{182}Au and ^{178}Pt , respectively [98, 99, 102, 103, 104]. (*: energy determined from α - γ coincidences)

the βDF partial half-life [105] $T_{1/2p,\beta\text{DF}} > 5.2 \times 10^7$ s is extracted and the upper limit for the βDF probability [105] is given by $P_{\beta\text{DF}} < 3.4 \times 10^{-8}$. These values are also the edge within which 95% of the events will be found. From the systematic behavior of $T_{1/2p,\beta\text{DF}}$ with respect to the difference between the parent Q-value for β decay Q_β and the daughter fission barrier B_f , $Q_\beta - B_f$, presented in [106], a $T_{1/2p,\beta\text{DF}}$ value in the order of 10^5 - 10^6 s is expected for ^{182}Tl . The experimental lower limit for $T_{1/2p,\beta\text{DF}}$ in the βDF decay of ^{182}Tl is thus more than two orders of magnitude larger than the estimated values from systematics [106]. However, it should be noted that $T_{1/2p,\beta\text{DF}}$ and $P_{\beta\text{DF}}$ are calculated, similar to the α -branching ratio, using the 351 keV $2^+ \rightarrow 0^+$ transition in ^{182}Hg which contains both low-and high-spin contributions. Consequently, the lower and upper limit for $T_{1/2p,\beta\text{DF}}$ and $P_{\beta\text{DF}}$ respectively, are only overall values since it is not possible to differentiate between the two

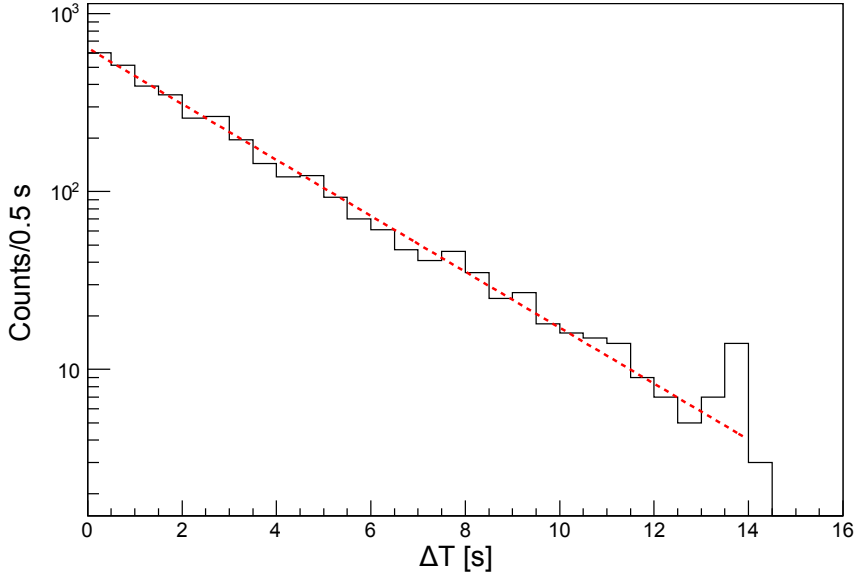


Figure 4.20: [Color] Time distribution of the events in the 6360 and 6406 keV ¹⁸²Tl α lines, collected at the decay position (Si3 and Si4). The dotted red line shows the result of a fit by an exponential curve resulting in the half-life value of 1.9(1) s.

isomers. Further experimental work to investigate these findings by producing isomerically purified radioactive ion beams is underway.

Figure 4.21a) shows a prompt α - γ coincidence matrix for ¹⁸²Tl while the corresponding projections on the γ -energy axis are given in spectra 4.21b), 4.21c) and 4.21d). We can identify different groups of α - γ coincident events using a prompt timing window of 500 ns. An overview of the observed α decays and coincident γ rays for ¹⁸²Tl is given in Table 4.4. No coincidence relation between the two α lines at 6406 and 6360 keV and any γ rays is observed (see Fig. 4.21a)). However, the difference in Q_α between these α decays is 46 keV which is below the detection limit of our HPGe detectors. Due to the complexity of the α -decay pattern of this odd-odd nucleus and limited knowledge about the structure of daughter isotope ¹⁷⁸Au, only a limited α -decay scheme can be constructed (see discussion).

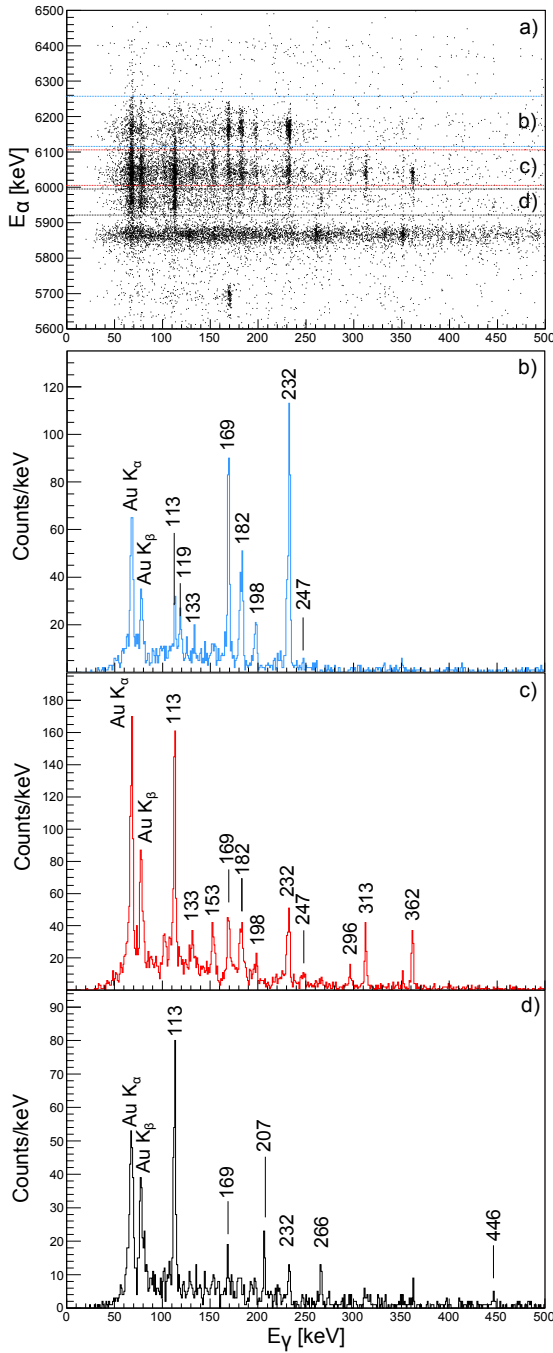


Figure 4.21: [Color] a) the prompt α - γ coincidence matrix for mass 182. b), c) and d) projections on the γ -energy axis for the events between the horizontal dashed lines corresponding to the different α - γ groups in spectrum a) (see also Fig. 4.19). The energy of the coincident γ rays is indicated in keV. The events in spectrum a) around 5700 keV are from fine structure in the α decay of ^{182}Hg .

Table 4.4: Overview of ¹⁸²Tl low-spin α decay, including the α -decay branching ratio b_α , energy E_α , intensity I_α (Si2), relative intensity $I_{rel,\alpha}$ (Si2), reduced α -decay width δ_α^2 and γ rays observed in coincidence (Si2 with Ge1). The sum $Q_\alpha + E_\gamma$ is also given for a number of transitions. The uncertainties are indicated between brackets and include both statistical and systematic contributions.

b_α (%)	E_α (keV)	I_α (counts)	$I_{rel,\alpha}$ (%)	δ_α^2 (keV)	E_γ (keV)	$Q_\alpha + E_\gamma$ (keV)
>0.49	6406 ^a	9367(97)	21(3)	>0.017		6550
	6360(6)	7301(85)	16(3)	>0.019		6503(6)
	6165(6) ^b	28080(168)	62(10)	>0.45	247.2(5)	6551(6)
					232.2(1)	
					197.5(2)	6501(6)
					182.3(2)	
					169.2(1)	
					132.9(4)	
					118.7(3)	
					112.9(2)	
					Au K $_\beta$	
					Au K $_\alpha$	
	6046(5) ^b	45500(213)	100	>2.3	361.5(1)	6543(6)
					312.6(1)	
					296.7(3)	
					247.8(7)	
					231.8(2)	
					197.5(8)	
					182.7(3)	
					169.3(2)	
					153.4(2)	
					131.7(4)	
					112.9(1)	
					102.0(5)	
					Au K $_\beta$	
					Au K $_\alpha$	
	5962(5) ^b	20520(143)	45(7)	>2.4	446.1(14)	6542(5)
					265.8(2)	
					232.2(3)	
					206.7(1)	
					169.2(3)	
					112.9(1)	
					Au K $_\beta$	
					Au K $_\alpha$	

^a E_α used to calibrate the silicon detectors

^b E_α from α - γ coincidences

4.3.4 Discussion

The partial α -decay schemes depicted in Fig. 4.22 for the ^{184}Tl (10^-) and (7^+) states are constructed using the information from Table 4.3 and the excitation energy (506 keV) of the (10^-) intruder state relative to the (7^+) state [68]. The 6132 keV (10^-) α decay has a reduced α -decay width of 16(4) keV. The unhindered α decays in the odd-mass neighbors $^{183,185}\text{Tl}$ involving the $\pi 1h_{9/2}$ intruder orbitals have reduced α -decay widths of about 30 keV [76, 85, 23, 80]. The presumed intruder-based [$\pi 1h_{9/2} \otimes \nu 1i_{13/2}$] (10^-) state is thus α decaying moderately hindered by a factor of ~ 2 , relative to the unhindered α decays of neighboring $^{183,185}\text{Tl}$ isotopes. This hints to the influence of an extra unpaired neutron in the odd-odd ^{184}Tl case and indicates that the (10^-) α -decaying state in ^{184}Tl and the excited state at the level $\Delta + 206$ keV in Fig. 4.22, fed in ^{180}Au have a similar structure. Intruder states of $\pi 1h_{9/2}$ origin have been identified in heavier odd-odd $^{182,184,186}\text{Au}$, which supports also its possible presence in ^{180}Au . The state at $\Delta + 206$ keV in ^{180}Au , fed in the α decay of the (10^-) state, decays further by a number of γ rays of which the 206 keV is the highest in energy. As can be seen in Fig. 4.13a), the feeding α line of 6132 keV does not have an α - e^- tail, pointing to the existence of an isomeric level Δ fed by the 206 keV γ line. Based on the α - γ data of the (7^+) state, the level Δ cannot be the groundstate (see below). As mentioned above, the other four γ rays are seen in prompt coincidence with the summed α - e^- tail and are thus most probably decaying as part of different cascades from the level $\Delta + 206$ keV. A possible crossover α -decay branch from the (10^-) to the state labeled Δ in Fig. 4.22 is not observed, which leads to an upper limit of its reduced α -decay width of 0.014 keV. The presumed regular [$\pi 3s_{1/2} \otimes \nu 1i_{13/2}$] (7^+) decays with a reduced α -decay width of 4.9(12) keV to an excited state in ^{180}Au , labeled $\Delta + 183$ in Fig. 4.22, which further decays by a number of γ transitions from which the 363 keV is the highest in energy. This points to the existence of a number of levels below the level labeled Δ . However, no further conclusions can be drawn with respect to decays to the ^{180}Au ground state.

In Fig. 4.23 the partial α -decay scheme for the ^{184}Tl (2^-) state is given. The reduced α -decay widths range from 0.09 to 2.3 keV, indicating that all α -decay branches of this isomeric state are hindered. The 6161 keV transition is the α line with the highest energy in the decay of the (2^-) state. However, α - γ coincidences establish a level, labeled x, 17.1(3) keV lower than the level fed by the 6161 keV line and this α decay is thus not the crossover transition. The 6161 keV α line is broader at the right-hand-side relative to the ^{184}Hg α -decay peak, which indicates also here α - e^- summing. This can be considered as additional evidence for the level at 17.1(3) keV, since the conversion electrons depopulating this level may sum up with the 6161 keV α line if both are

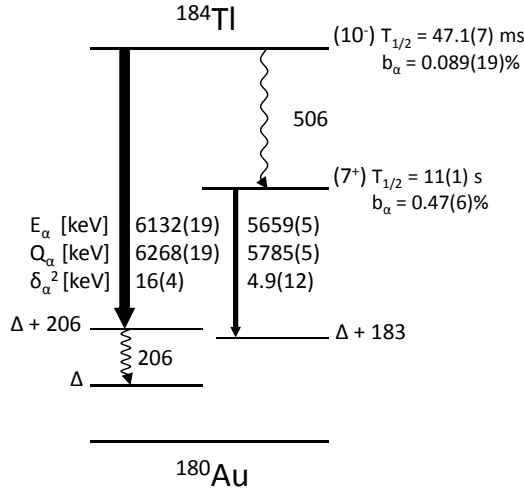


Figure 4.22: Partial α -decay scheme for the (10^-) and (7^+) states in ¹⁸⁴Tl. The E_α , Q_α values and corresponding reduced α -decay widths δ_α^2 are indicated next to the corresponding α decays, together with the energy of selected observed coincident γ rays.

registered simultaneously in the same silicon detector. It cannot be concluded that the level x is the ground state of ¹⁸⁰Au. Combining the information of the three α -decaying states in ¹⁸⁴Tl, there is evidence of at least two long-living states (the states x and Δ) in ¹⁸⁰Au.

In the partial α -decay scheme of ¹⁸²Tl in Fig. 4.24 one can distinguish three groups of decays, based on their similar reduced α -decay widths. It should be noted that, since the α -decay branching ratio of 0.49% is a lower limit, the reduced α -decay widths are lower limits as well. The α -decay transitions with energies of 5962 and 6046 keV both have reduced α -decay widths of about 2.5 keV. This points to a similar structure of the two excited levels at 446 and 362 keV in daughter ¹⁷⁸Au. The α decay of 6165 keV feeding a level at 247 keV has a reduced α -decay widths of 0.45 keV, a factor of ~ 5 more hindered compared to the 5962 and 6046 keV α decays. However, the 6360 keV and full-energy 6406 keV transitions are much more hindered ($\delta^2 \approx 0.02$ keV), a factor of 130 relative to the 5962 and 6046 keV α decays and 25 relative to the 6165 keV. The similar reduced widths of the strongly hindered transitions to the (supposed) ground state and excited level at 46 keV indicate also to the similar structure of both levels.

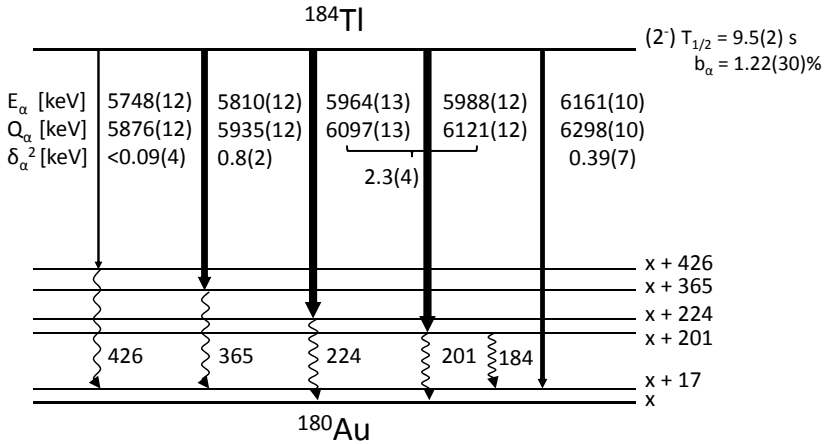


Figure 4.23: Partial α -decay scheme for the (2^-) state in ^{184}Tl . The E_α , Q_α values and corresponding reduced α -decay widths δ_α^2 are indicated next to the corresponding α decays, together with the energy of selected observed coincident γ rays.

The hyperfine structure pattern of the α -decaying state in ^{182}Tl is similar to that of ^{180}Tl [84], for which a spin assignment $I = 4, 5$ was proposed in [26]. Based on this argument and the close values of the corresponding magnetic moments, it is tempting to assign to these states the same spin and configuration [84]. As mentioned in the introduction, a neutron configuration change to $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]_{4^-, 5^-}$ can be expected to occur around $^{180, 182}\text{Tl}$. Long-living states with this assumed structure have been identified in $^{178, 180}\text{Tl}$ [107, 108]. In the α decay from this state in ^{180}Tl , five α lines have been observed by Toth et al [108] and recently also by Kondev et al. [109]. The two lowest energy α transitions, 6208 and 6281 keV, have reduced α -decay widths of 4.9(36) and 4.2(29) keV respectively, which corresponds remarkably well to the situation in ^{182}Tl , confirming the probably identical structures of the parent states in both isotopes. Also in ^{178}Tl , the two α lines with the lowest energy values, decay unhindered to excited levels in ^{174}Au [107]. The broad range in reduced α -decay widths from almost no hindrance to a hindrance of about 2000 relative to odd-thallium neighbors, which could be determined because of the high statistics obtained, points to strong structural changes between parent thallium and daughter gold isotopes. ^{182}Tl is characterized by rather pure nearly spherical states involving proton-holes in the $Z = 82$ shell, while the respective gold isotope is strongly deformed [110].

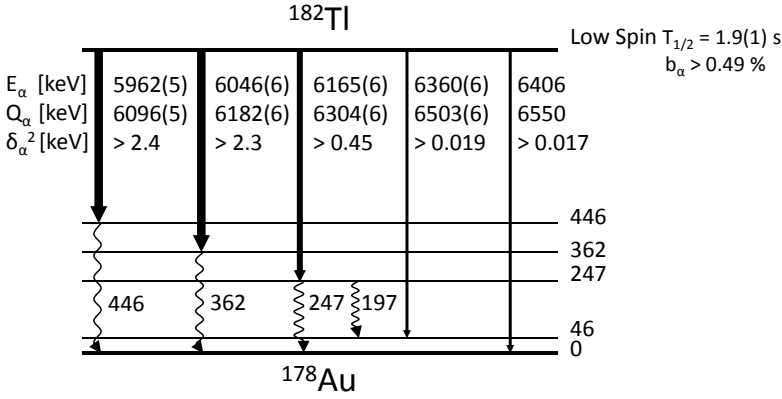


Figure 4.24: Partial α -decay scheme for the low-spin state in ^{182}Tl . The E_α , Q_α values and corresponding reduced α -decay widths δ_α^2 are indicated next to the corresponding α decays, together with the energy of observed coincident γ rays.

4.3.5 Conclusion

The α decay of $^{182,184}\text{Tl}$ was studied at the ISOLDE facility. The α decay of the three long-lived states in ^{184}Tl , (10^-) , (7^+) and (2^-) , has been disentangled and for the first time, α -decay branches for the (10^-) and (7^+) states were identified. Due to the purity of the singles α -decay energy spectra for all three states, α -decay branching ratios of 0.089(19)%, 0.047(6)% and 1.22(30)% were extracted for the (10^-) , (7^+) and (2^-) states respectively. A more precise half-life of 9.5(2) s was deduced for the (2^-) state. New α -decay lines were identified also for ^{182}Tl , all of them have a half-life of 1.9(1) s. A lower limit of the α -decay branching ratio of 0.49% could be determined for this isotope. Detailed α - γ coincidence analysis was performed, however only partial α -decay schemes could be constructed for $^{182,184}\text{Tl}$ due to the complexity of the α -decay patterns, limited knowledge on the structure of the daughter nuclei and lack of γ - γ coincidences. The combination of low-lying states in ^{178}Au identified in this work through α - γ coincidences, the laser spectroscopy studies at ISOLDE and the data obtained at a recently performed in-beam study of ^{178}Au at RITU should help to establish the full decay scheme of ^{178}Au . A high-spin and low-spin α -decaying state have already been identified by our latest laser spectroscopy studies at ISOLDE [111]. Reduced α -decay widths or hindrance factors have been calculated for the different α -decay branches in $^{182,184}\text{Tl}$. From a comparison with values obtained in neighboring odd-A and even-A

thallium isotopes, analogies were deduced between states with possibly similar or completely different structures. Only the ^{184}Tl (10^-) state is α decaying unhindered to a state in ^{180}Au , all other α -decay channels observed in this study are hindered, indicating a different underlying structure of the mother and daughter isotopes.

4.3.6 Acknowledgements

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5 | Laser Spectroscopy of $^{179-184}\text{Tl}$

In this chapter the experimental results of the laser spectroscopy study of $^{179-184}\text{Tl}$ are presented. The results presented are obtained from measurements at the ISOLDE facility. In the first section, the data taking and extraction are described, followed by an overview of the hyperfine spectra of all masses, including a fit of the data in the second section. A more detailed discussion of the spin assignment and possible configuration mixing in $^{180,182,184}\text{Tl}$ and the unexpected increase in the isomer shift in ^{183}Tl is discussed in the last section. The results of the other isotopes are included for completeness.

5.1 Extraction of hyperfine spectra and fitting procedure

As mentioned in chapter 3, the excitation from the atomic ground state $6p^2P_{1/2}$ to an intermediate electronic state $6d^2D_{3/2}$ (36117.9 cm^{-1}) was performed by a frequency-doubled narrow-band tunable dye laser beam at 276.9 nm with a linewidth of 0.8 GHz. The subsequent ionization of excited thallium atoms was accomplished by the 532 nm laser beam from the 10 kHz repetition rate EdgeWave Nd:YAG laser, that was also used as a pump source for the first excitation step laser (for details of RILIS laser setup see [31]).

During a laser scan, the measurement time for each laser frequency was one supercycle, consisting of a fixed number of proton pulses. Throughout the experiment, the composition of the supercycle was not identical for the different scans. The number of proton pulses per supercycle sent to ISOLDE varied between 15 and 20. The selected frequency difference between two consecutive

steps varied between 150 to 300 MHz. In the hyperfine spectra, the intensity is given as a function of laser frequency for specific γ lines or α particles, characteristic for the isotope of interest. The fitting of the hyperfine spectra is done using Voigt profiles [112], including 8 parameters. The parameters are the spin, isotope shift, A and B constants, the Lorentz and Doppler widths, the amplitude of the peaks and the possible constant or linearly varying background. The results presented in this work did not take into account the possible variation in the laser power and proton intensity, saturation and polarization effects. The correction for these effects is not straightforward and a more sophisticated fitting procedure is carried out by Maxim Seliverstov, including these four effects. However, in the case of thallium, the effects are subtle and the conclusions presented in this work remain the same.

To calculate the magnetic moments of the thallium isotopes, equation (2.31) is used with $\mu_{ref}({}^{205}\text{Tl}) = 1.6382136(1)$ n.m. [113] and $A_{ref}({}^{205}\text{Tl}, 6p^2P_{1/2}) = 21310.835(5)$ MHz [114].

The changes in the mean-square charge radii $\delta\langle r^2 \rangle^{A,A'}$ are deduced from the measured isotope shift $\delta\nu^{A,A'}$ by the following relation [59]:

$$\delta\nu^{A,A'} = M \frac{A - A'}{AA'} + F\lambda^{A,A'} \quad (5.1)$$

where M includes both the normal and specific mass shift constants and $\lambda^{A,A'} = K(Z) \delta\langle r^2 \rangle^{A,A'}$. $K(Z)$ takes into account the contribution of higher-order radii moments in $\lambda^{A,A'}$ (see Refs. [7, 59, 60]). In this work, the value $K(81) = 0.938$ is used [7]. For the F and M constants, the values deduced in Ref. [7] are used: $F(276.9 \text{ nm}) = 10.3788 \text{ GHz fm}^{-2}$ and $M(276.9 \text{ nm}) = 701(330) \text{ GHz amu}$.

For a more detailed description of hyperfine spectra and the data extraction, we refer the reader to literature, e.g. [36].

5.2 Hyperfine spectra of ^{179–184}Tl

In this section, an overview is given of the hyperfine spectra of ^{179–184}Tl to illustrate the quality of the collected data (see Fig. 5.1-5.10). It should be noted that the frequencies indicated in the plots are the values before frequency-doubling. The spins indicated in the different spectra are tentative assignments based on a combination of systematics and results from decay spectroscopy studies. However, these hyperfine spectra can be fitted also with other spin

values (see further). This is not a matter of the resolution, but the consequence of the chosen transition with electronic spin $1/2$ of the lower level and negligible splitting of the upper level. Figure 5.10 shows a summary of all spectra and the center of gravity is indicated by a vertical dashed line in each hyperfine spectrum. The γ or α line(s), possibly from daughter isotopes, used to produce the hyperfine spectra are noted in the different spectra. In appendix E an overview is given of the collected singles α -decay spectra of $^{179-184}\text{Tl}$.

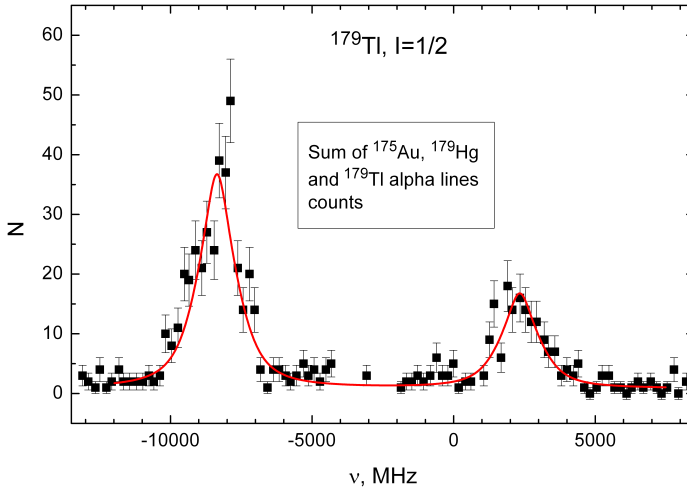


Figure 5.1: [Color] Hyperfine spectrum of ^{179}Tl , using the α decays of ^{179}Tl , ^{179}Hg and ^{175}Au . The red line results from a fit of the spectrum, assuming $I = 1/2$.

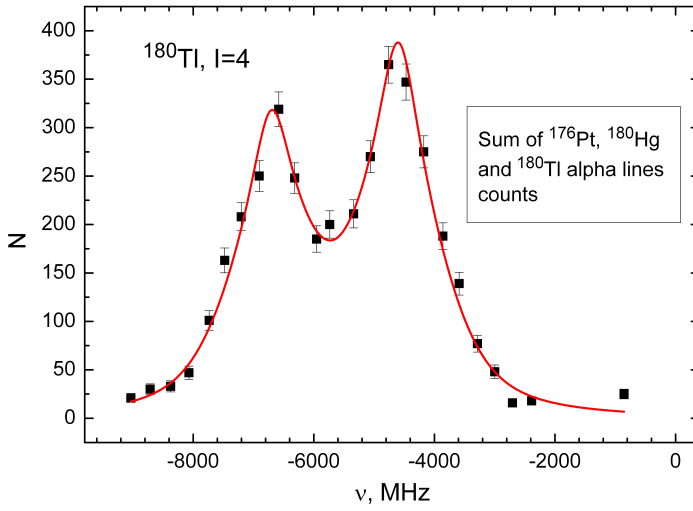


Figure 5.2: [Color] Hyperfine spectrum of ^{180}Tl , using the α decays of ^{180}Tl , ^{180}Hg and ^{176}Pt . The red line results from a fit of the spectrum, assuming $I = 4$.

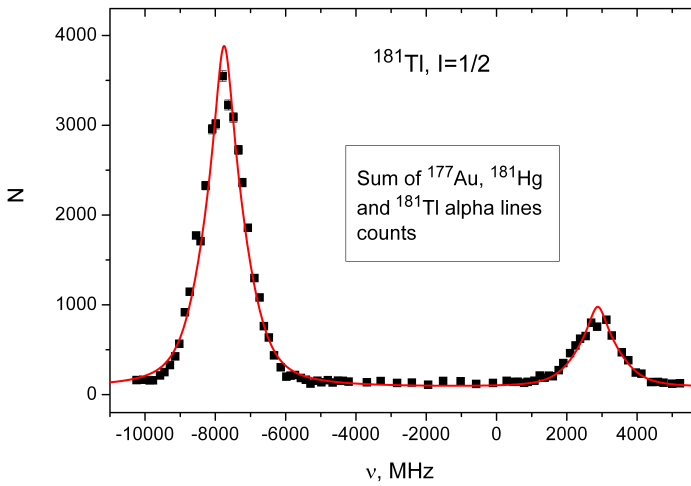


Figure 5.3: [Color] Hyperfine spectrum of ^{181}Tl , using the α decays of ^{181}Tl , ^{181}Hg and ^{177}Au . The red line results from a fit of the spectrum, assuming $I = 1/2$.

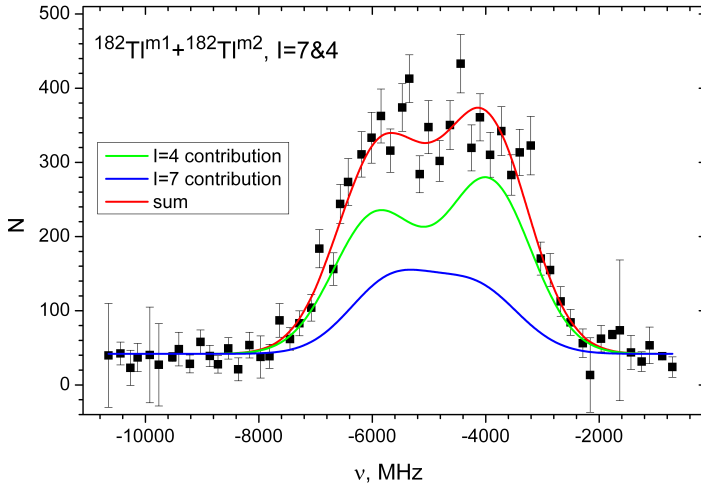


Figure 5.4: [Color] Hyperfine spectrum of ^{182}Tl , using the $6^+ \rightarrow 4^+$ (332 keV) transition in ^{182}Hg . The different lines result from fits of the spectrum, including contributions from low-spin $I = 4$ (green) and high-spin $I = 7$ (blue) states, as well as the sum of both (red). See section 5.4.4 for a more detailed discussion.

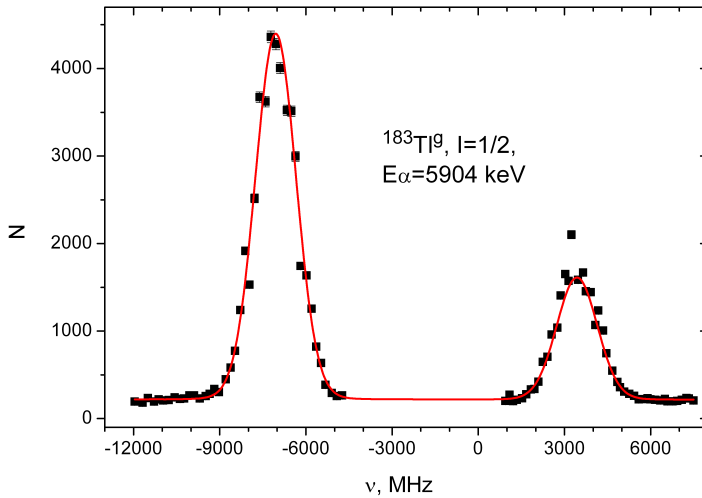


Figure 5.5: [Color] Hyperfine spectrum of ^{183g}Tl , using the 5904 keV α decay of ^{183}Hg . The red line results from a fit of the spectrum, assuming $I = 1/2$.

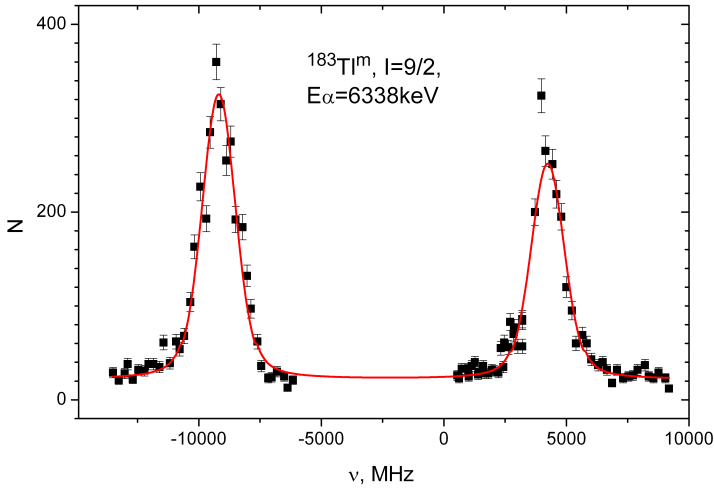


Figure 5.6: [Color] Hyperfine spectrum of ^{183m}Tl , using the 6338 keV α decay of ^{183m}Tl . The red line results from a fit of the spectrum, assuming $I = 9/2$.

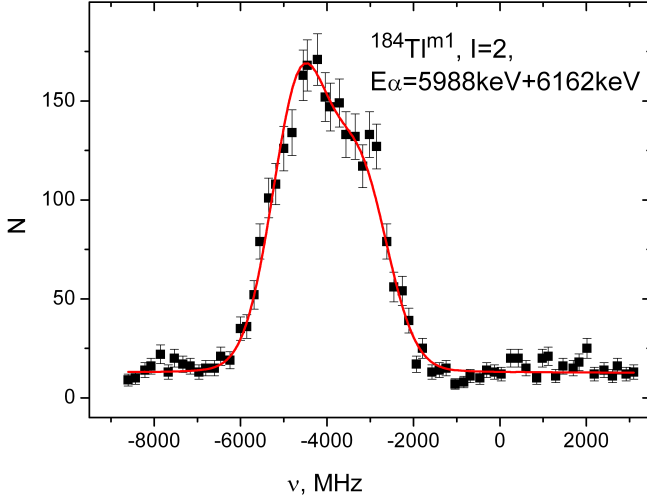


Figure 5.7: [Color] Hyperfine spectrum of $^{184m1}\text{Tl}$, using the 5988 and 6162 keV α decays of $^{184m1}\text{Tl}$. The red line results from a fit of the spectrum, assuming $I = 2$.

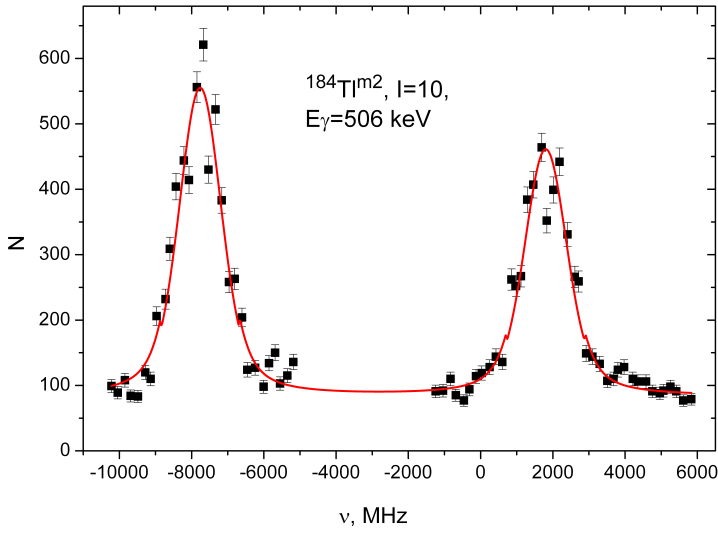


Figure 5.8: [Color] Hyperfine spectrum of $^{184m2}\text{Tl}$, using the 506 keV IT of $^{184m2}\text{Tl}$. The red line results from a fit of the spectrum, assuming $I = 10$.

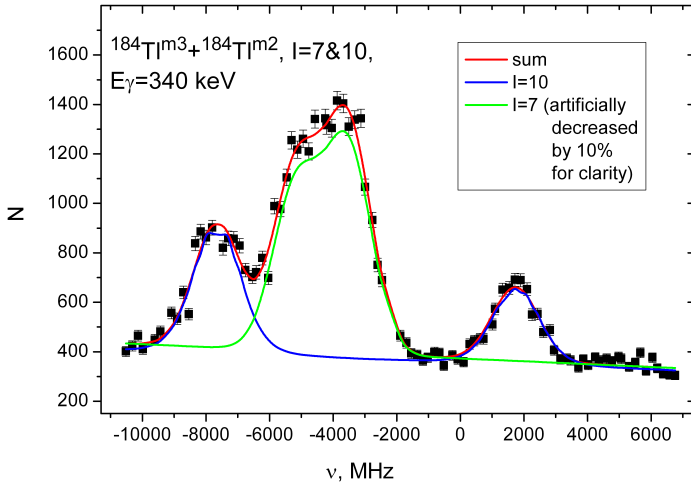


Figure 5.9: [Color] Hyperfine spectrum of $^{184m2,m3}\text{Tl}$, using the 340 keV ($6^+ \rightarrow 4^+$) transition in ^{184}Hg . The different lines result from fits of the spectrum, assuming $I = 7$ (green) and $I = 10$ (blue), as well as the sum of both (red).

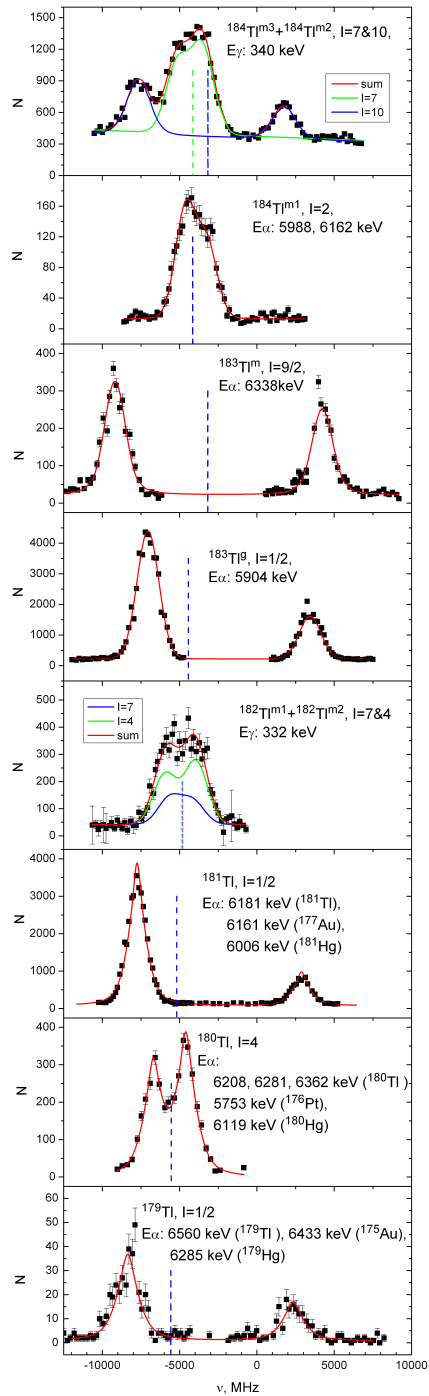


Figure 5.10: [Color] Overview of the hyperfine spectra of $^{179-184}\text{Tl}$, including fits of the different spectra. The center of gravity of the atomic levels is indicated for each hyperfine spectrum with a vertical blue line.

5.3 Overview of the results for $^{179-184}\text{Tl}$

In Table 5.1, an overview of the laser spectroscopy results for $^{179-184}\text{Tl}$ obtained in this work is given. It should be noted that the quadrupole constant is small in comparison with our experimental uncertainties (see also [7]). The constant B was varied during the fitting, however this did not influence the results and it was fixed to zero for the final fits.

Table 5.1: Overview of the laser spectroscopy results for $^{179-184}\text{Tl}$ from ISOLDE. The results from IRIS are added for comparison.

A	I	IS ^c (Mhz)	A (Mhz)	$\delta\langle r^2 \rangle^c$ (fm ²)	μ (μ_N)	$\delta\langle r^2 \rangle^d$ (fm ²)	μ^d (μ_N)
184	2	-9910(300)	1045(110)	-0.98(3)	0.32(3)	-0.995(30)	0.316(99)
184	7	-9880(220)	399(18)	-0.97(2)	0.43(2)		
184	10	-7950(150)	1820(50)	-0.78(2)	2.80(8)		
183	9/2	-7970(160)	5290(80)	-0.78(2)	3.66(6)		
183	1/2	-10470(150)	20868(90)	-1.03(2)	1.60(1)	-1.056(28)	1.618(37)
182	7 ^a	-11320(220)		-1.12(2)			
182	4	-11290(210)	-870(40)	-1.12(2)	-0.54(3)		
181	1/2	-11980(250)	21313(200)	-1.18(3)	1.64(2)		
180	4 ^b	-12660(150)	-943(24)	-1.25(2)	-0.58(2)		
180	5 ^b	-12690(160)	-769(26)	-1.26(2)	-0.59(2)		
179	1/2	-13050(320)	21371(280)	-1.29(3)	1.64(2)		

^aA parameter fixed to 399 MHz in fitting procedure

^bResults from a fit of the same hyperfine spectrum assuming spin I = 4 or 5

^cRelative to ^{205}Tl

^dResults from IRIS (Gatchina) [7]

5.4 Spins and moments of $^{179-184}\text{Tl}$

In Fig. 5.11 the magnetic moments of the different thallium nuclear states are presented, where the full symbols represent (older) literature data and the hollow and semi-hollow symbols represent data measured at the IRIS and ISOLDE facilities, respectively. The data obtained at the IRIS facility have been published recently in [7]. The newly measured magnetic moments follow the isotopic trend for the lighter or heavier nuclei for some of the spins fairly well. This points to the smooth change in the nuclear structure of the thallium nuclei around neutron midshell ($N = 104$) and beyond.

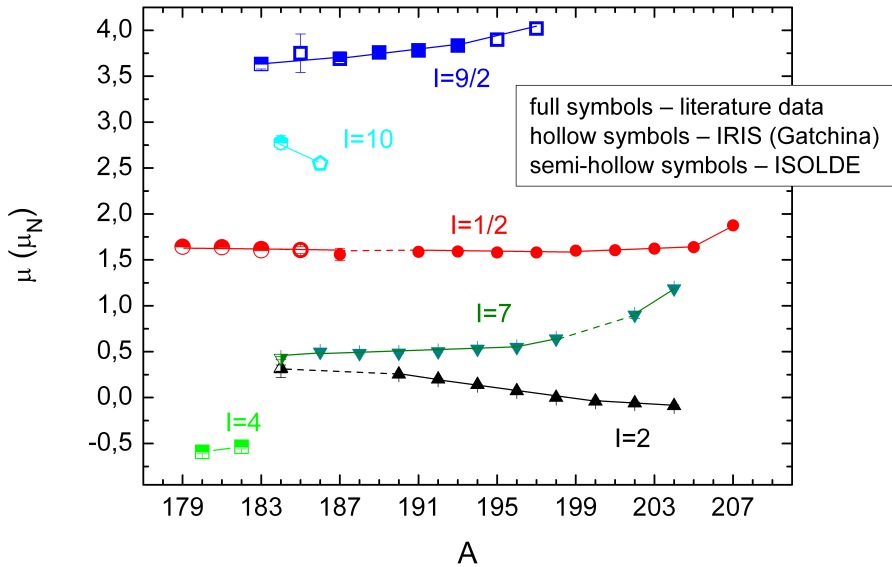


Figure 5.11: [Color] Isotopic dependence of magnetic moments of the different thallium nuclear states. The IRIS (Gatchina) data are from Ref. [7].

5.4.1 Spin assignments in ^{184}Tl

The hyperfine spectra in Fig. 5.7, 5.8 and 5.9 can also be fitted with other spin assignments. However, this leads to a noticeable discrepancy between the calculated magnetic moments, using the additivity relation (equation (2.33)) and the measured ones (see e.g. Table 5.2). The spins assigned to the long-lived states in ^{184}Tl , (10^-) , (2^-) and (7^+) , remain tentative and are based on systematics and previous and present decay studies. One can report that with these assignments, the magnetic moments for the $I = 2$ and $I = 7$ states fit the systematics for similar states in neighboring thallium isotopes, see Fig. 5.11. From the present decay study, reported in chapter 4, it is clear that the spin difference between the two high-spin states ((10^-) and (7^+)) is $3\hbar$ and that the two states have opposite parity. The spin assignment of the (10^-) intruder state in ^{184}Tl is discussed in more detail in the next section.

Spin assignment of the (10^-) state in ^{184}Tl

The $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ configuration has been proposed by Andreyev et al. [71] for the proton intruder state in ^{184}Tl . In chapter 4 evidence is presented that supports the interpretation of the intruder character. A spin $I = 10$ was assigned to this state based on α -decay energy systematics and hindrance factors [71]. The quality of the fit of the hyperfine spectrum of ^{184}Tl with the assumption of spin $I = 8, 9, 10$ or 11 is the same. However, comparison of the experimental and calculated magnetic moments using the additivity relation (equation (2.33)) can discriminate between the different spins. For the $\pi 1h_{9/2}$ configuration, the measured magnetic moment $\mu(\pi 1h_{9/2}) = 3.75(21)$ n.m. from the neighboring ^{185}Tl [7] can be used in the calculations. For the $\nu 1i_{13/2}$ configuration the magnetic moments involving this configuration in the odd-neutron ^{185}Hg , ^{185}Pb and ^{193}Po isotopes seem appropriate. However, during recent isotope shift and hyperfine structure measurements for a long chain of polonium isotopes [36], it was shown that $\mu(\nu 1i_{13/2})$ grows rather rapidly when the deformation increases. In that respect, it would be misleading to use $\mu(\nu 1i_{13/2})$ of ^{185}Hg or ^{185}Pb since these isotopes have a mean-square deformation of only $\langle \beta^2 \rangle^{1/2} (^{185}\text{Hg}) = 0.16(1)$ [115] and $\langle \beta^2 \rangle^{1/2} (^{185}\text{Pb}) \approx 0.1$ [21], both markedly lower than the mean-square deformation of the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ intruder state in ^{184}Tl , $\langle \beta^2 \rangle^{1/2} (^{184}\text{Tl}, I = 10) = 0.21(2)$ (see also Fig 1.4 and 5.17). However, the mean-square deformation $\langle \beta^2 \rangle^{1/2} (^{193}\text{Po}) = 0.22$ [36] is comparable to that of the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ configuration in ^{184}Tl and is thus preferable in the calculations.

The experimentally determined magnetic moments and the results of the calculations for the different spins are summarized in Table 5.2. From these calculations it is clear that spin assignments $I = 8, 9$ lead to a noticeable discrepancy between the calculated and measured magnetic moments. Both spin $I = 10$ and 11 cannot be excluded with purely additivity-rule considerations. However, based on the good correspondence between the calculated and measured magnetic moment for $I = 10$ and other spectroscopic results [105, 68] and theoretical predictions [12, 11], this spin assignment is strongly supported.

5.4.2 Configuration mixing of (7^+) state in ^{184}Tl

Figure 5.12 shows a detailed view of the magnetic moments for the states with spin $I = 7$ in the odd-odd thallium isotopes. The magnetic moment for the $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ using the additivity relation (equation (2.33)) coincides with the experimental results only for $A = 198$. The single-particle moment for the $\pi 3s_{1/2}$ configuration used in the calculations represents the approximate mean

Table 5.2: Measured and calculated magnetic moments for the $[\pi 1h_{9/2} \otimes \nu 1i_{13/2}]$ intruder configuration in ^{184}Tl , assuming different spins.

A	I	$\mu_{exp}(\mu_N)$	$\mu_{calc}(\mu_N)$
184	8	2.7(1)	1.6(2)
184	9	2.7(1)	2.1(2)
184	10	2.8(1)	2.6(3)
184	11	2.8(1)	3.0(3)

^aCalculation with additivity relation, using $\mu(\pi 1h_{9/2}, ^{185}\text{Tl}) = 3.75(21)$ [7] and $\mu(\nu 1i_{13/2}, ^{193}\text{Po}) = -0.741(65)$ [36]

value from the experimental magnetic moments of thallium ($\pi 3s_{1/2}$) in the mass region $A = 194 - 204$ [90]. For the $\nu 1i_{13/2}$ configuration, the approximate mean values of the single-particle moments of mercury or lead in the mass region $A = 194 - 204$ can be used, since the shape of the $I = 7$ state in ^{184}Tl has a near-spherical shape, comparable to that of mercury and lead [90]. The increase in the thallium magnetic moments for $A > 198$ was explained by a small admixture of $7^+ [\pi 1h_{11/2} \otimes \nu 3p_{3/2}]$ and $7^+ [\pi 1h_{11/2} \otimes \nu f_{5/2}]$ configurations which have a large positive magnetic moment [90]. From $A < 198$ on, the magnetic moments decrease by nearly 20% and stabilize at 0.5 n.m. for $A = 192-186$. This decrease may be explained by an admixture of the $7^+ [\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration with a negative magnetic moment of about 1 n.m. (using for the $\nu 1i_{13/2}$ configuration the approximate mean values of the single-particle moments of mercury or lead and for the $\pi 2d_{3/2}$ configuration the mean value of the single-particle moments of $^{203-205}\text{Tl}$ ($\pi 2d_{3/2}) = 0.29(5)$ n.m. [24]). This has also been pointed out by Ekstrom et al. [90]. For $A = 184$ one notices a further decrease. This can be related to an increase of the $7^+ [\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ admixture. In ^{184}Tl the retardation of the transition from the intruder-based (10^-) state to the (7_1^+) state is about seven times smaller than the equivalent transition in ^{186}Tl , see chapter 4. This also hints towards increased mixing of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration in the (7_1^+) state.

5.4.3 Configuration mixing of (2^-) state in ^{184}Tl

Ekstrom et al. [90] interpreted the spin $I = 2$ states in the doubly-odd $^{194-204}\text{Tl}$ isotopes as a mixture of the configurations $2^- [\pi 1s_{1/2} \otimes \nu 2f_{5/2}]$ and $2^- [\pi 1s_{1/2} \otimes \nu 3p_{3/2}]$, with a possible contribution from $2^- [\pi 2d_{3/2} \otimes \nu 3p_{1/2}]$.

Calculation of the magnetic moment of the (2^-) state in ^{184}Tl using the

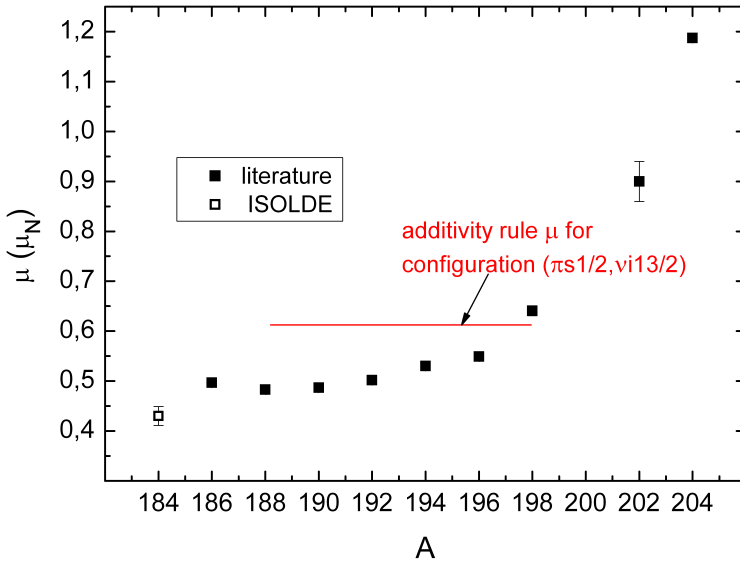


Figure 5.12: Detailed view of the isotopic dependence of the magnetic moments for the states with spin $I = 7$ in the odd-odd thallium isotopes.

additivity relation (equation (2.33)) for a $[\pi 1s_{1/2} \otimes \nu 3p_{3/2}]$ configuration leads to $\mu_{calc} = 0.51(2)$ n.m. This value is reasonably close to the experimental value $\mu_{exp} = 0.32(3)$ n.m. In the calculation, the magnetic moments of ^{185}Tl [7] and ^{185}Pb [116] were used for the proton and neutron configurations respectively. Nevertheless, the possibility of some admixture of the $[\pi 2d_{3/2} \otimes \nu 3p_{1/2}]$ configuration cannot be excluded. For ^{184}Tl a neutron configuration based on the $f_{5/2}$ shell may be excluded as it is expected to be at a much higher excitation energy in thallium isotopes close to the neutron midshell [71]. An overview of the calculated magnetic moments assuming different configurations is given in Table 5.3.

As can be seen in Fig. 5.11, the magnetic moments of the $I = 2$ states slowly increase when going to the more neutron deficient side. The reason for this increasing trend is the decreasing admixture of the $[\pi 1s_{1/2} \otimes \nu 2f_{5/2}]$ configuration with decreasing neutron number.

Table 5.3: Measured and calculated magnetic moments for the (2^-) state in ^{184}Tl , assuming different configurations.

A	I	$\mu_{exp}(\mu_N)$	$[j_p \otimes j_n]$	$\mu_{calc}(\mu_N)$
184	2	0.32(3)	$[\pi 1s_{1/2} \otimes \nu 3p_{3/2}]$	0.51(2) ^a
184	2	0.32(3)	$[\pi 1s_{1/2} \otimes \nu 2f_{5/2}]$	-0.27(1) ^b
184	2	0.32(3)	$[\pi 1s_{1/2} \otimes \nu 2f_{5/2}]$	-0.44(1) ^c
184	2	0.32(3)	$[\pi 2d_{3/2} \otimes \nu 3p_{1/2}]$	0.80(14) ^d

^aCalculation with additivity relation, using the magnetic moments of ^{185}Tl [7] and ^{185}Pb [116] for the proton and neutron configurations respectively

^bCalculation with additivity relation, using the magnetic moments of ^{185}Tl [7] and ^{197}Hg [116] for the proton and neutron configurations respectively

^cCalculation with additivity relation, using the magnetic moments of ^{185}Tl [7] and ^{201}Pb [116] for the proton and neutron configurations respectively

^dCalculation with additivity relation, using the magnetic moments of $^{203-205}\text{Tl}$ (mean value) [116] and ^{185}Hg [116] for the proton and neutron configurations respectively

5.4.4 Spin assignment in ^{182}Tl

In accordance with the results of the α -decay study of ^{186}Bi [70], clear evidence was found for the presence of two isomeric states in ^{182}Tl from β -decay studies [65]. The hyperfine structure patterns for different α and γ lines in the ^{182}Tl decay spectra, obtained in narrow-band laser scans, support this observation.

The hyperfine structure pattern of the $8^+ \rightarrow 6^+$ (413 keV) and $6^+ \rightarrow 4^+$ (332 keV) γ transitions in β -decay daughter ^{182}Hg , as a function of laser frequency is distinctly different from the pattern of the $2^+ \rightarrow 0^+$ (351 keV) transition, indicating a high-spin β -decaying state and a low-spin β -decaying state. This is illustrated in Fig. 5.13 a) and b). The difference in the hyperfine patterns of both presents itself in the dip visible in the spectra resulting from the low- and high-spin contributions and the absence of this dip in the spectra with increased contribution from the high-spin state. Figure 5.13 c) shows the hyperfine spectrum of the 511 keV γ line. The hyperfine structure pattern of the 511 keV γ line is similar to that of the $2^+ \rightarrow 0^+$ (351 keV) transition and thus also dominated by the contribution of the low-spin state. Two frequency ranges, labeled A and B, are indicated in Fig. 5.13 with vertical dashed lines. These frequency ranges A and B were selected to characterize both the low- and high-spin states and the low-spin state respectively and will be used further in this section.

All α lines in our study have a hyperfine structure pattern testifying to the

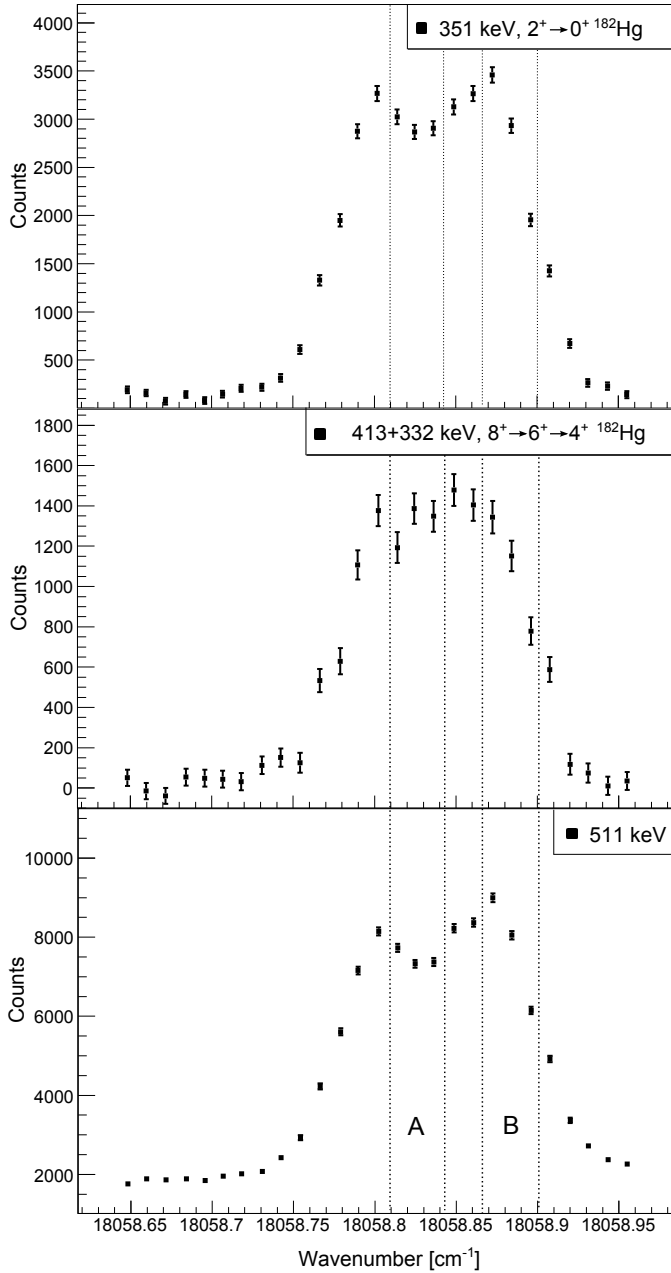


Figure 5.13: Intensity pattern of the $2^+ \rightarrow 0^+$ (351 keV) (a), $8^+ \rightarrow 6^+$ (413 keV), $6^+ \rightarrow 4^+$ (332 keV) (b) γ transitions in ^{182}Hg and 511 keV (c) γ line, as a function of the first excitation step laser wave number, collected at mass 182. The difference in the intensity patterns indicate contributions from two isomeric states in ^{182}Tl (see text). Two frequency ranges, labeled A and B, are indicated with vertical dashed lines.

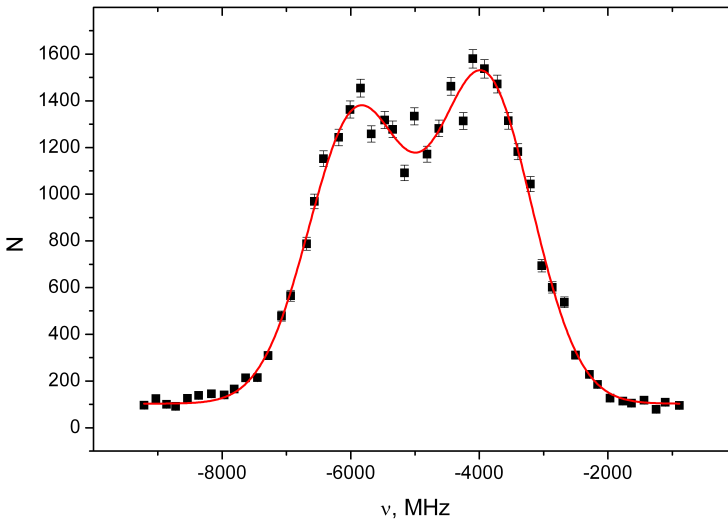


Figure 5.14: Intensity pattern of the ^{182}Tl α decays (6000-6430 keV) as a function of the first excitation step laser wave number. The red line results from a fit of the spectrum, assuming $I = 4$.

predominance of the low-spin state decay in these lines. A fit of the hyperfine spectrum of the ^{182}Tl α lines is shown in Fig. 5.14. In the fitting procedure, a spin $I = 4$ was assumed, based on the observed $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]$ configuration with $I = 4$ in ^{180}Tl and the similarities between the hyperfine spectra (see Fig. 5.2 and the next section).

A fit of the hyperfine spectrum of the $6^+ \rightarrow 4^+$ (332 keV) transition in ^{182}Hg , populated in the β decay of ^{182}Tl , is shown in Fig. 5.4. This spectrum is fitted assuming two contributions, one from a spin $I = 4$ and one from a spin $I = 7$. The tentative $I = 7$ assignment is based on the observed β -decay feeding, similarity with the hyperfine structure pattern and expected similar configuration of the $I = 7$ state in ^{184}Tl . Using only one contribution from a spin $I = 7$ in the fitting procedure leads to unreasonable results.

In the double fit of the ^{182}Tl hyperfine spectrum, the Lorentz and Gaussian widths of the Voigt profile are fixed for both isomers in accordance with the results obtained from a fit of the hyperfine spectrum of the α -decay lines assuming a spin $I = 4$, see Fig. 5.14. The magnetic moment and the center of gravity for the $I = 4$ contribution, extracted from the hyperfine spectrum of the α -decay lines, were also fixed in the double fit. The magnetic moment of the $I = 7$ contribution is also fixed during the fitting, using the value extracted

from the $I = 7$ state in ^{184}Tl . Fixing also the magnetic moment of the $I = 7$ contribution is preferred since the parameters are highly correlated and it is impossible to determine the magnetic moment for the $I = 7$ directly with this spectrum.

Figure 5.15 shows two α spectra A and B, where the frequencies of the laser have selected values. These values are indicated with the vertical dashed lines in Fig. 5.13 and correspond to frequencies inside the dip (A) and outside the dip (B). As mentioned above, the frequency ranges A and B were selected to characterize both the low- and high-spin states and the low-spin state respectively. Both α spectra are normalized to the intensity of the 5867 keV ^{182}Hg α line. Around 6250 keV, the intensity of spectrum A (blue) is higher relative to that of spectrum B (grey). This observation indicates that, out of comparison of these two α -decay spectra, there is evidence for α decay from the high-spin state in the region around 6250 keV. Similarly to ^{184}Tl it seems that the high-spin state has a considerably lower α -decay probability than the low-spin state. Unfortunately, no further information can be extracted.

The same method can be used to produce γ -ray spectra and two such frequency-gated γ -ray spectra are shown in Fig 5.16 for the frequency ranges labeled A and B in Fig. 5.13. The spectra are normalized to the intensity of the $2^+ \rightarrow 0^+$ (351 keV) transition in ^{182}Hg . In the spectra, the relative intensity of the different transitions is a measure for the feeding in the low- or high-spin β decay: the transitions where the intensity of spectrum A (blue) is higher relative to that of spectrum B (grey) are dominated by the high-spin β decay and vice versa. However, the discrimination power is too low to disentangle the β -feeding pattern of the two states and only a mixed β -decay scheme could be deduced [65].

To summarize, all hyperfine spectra produced by different γ or α -lines, may be consistently fitted with the assumption of the presence of two long-lived states in ^{182}Tl with different contributions of each state in the resulting hyperfine structure depending on the details of the α and β decay. Tentative $I = 7$ assignment for the high-spin state is in agreement with the fitting results. The sign and the value of the magnetic moment of the low-spin state ($-0.54(3)\text{n.m.}$) deduced with the assumption $I = 4$, point to the similarity of the configuration for the low-spin state of ^{182}Tl and ^{180}Tl (see below).

5.4.5 Spin assignment in ^{180}Tl

The $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]_{4^-,5^-}$ configuration has been proposed by Elseviers et al. [26] for the ground state of ^{180}Tl in a previous β -decay study at ISOLDE, CERN. The quality of the fit of the hyperfine spectrum of ^{180}Tl with the

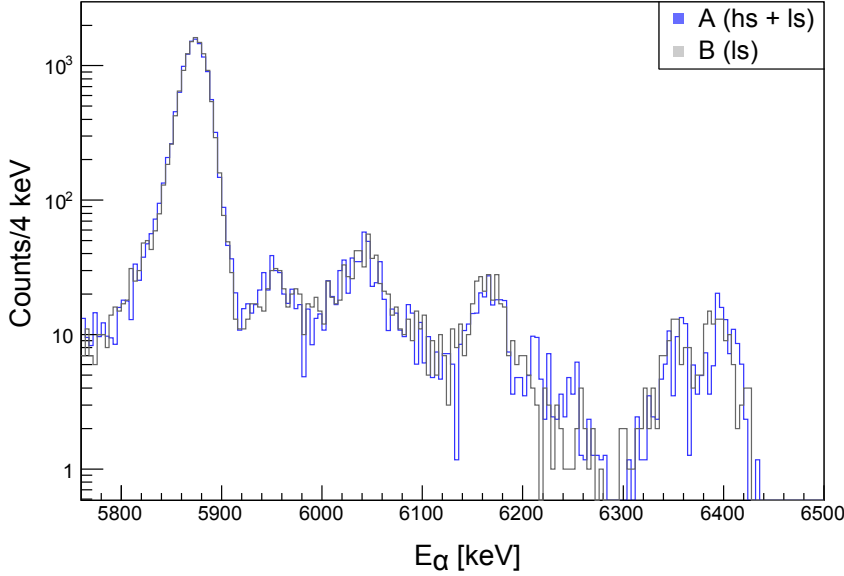


Figure 5.15: [Color] α spectra for the frequency range A (blue) and B (grey) collected at mass 182.

assumption of spin $I = 4$ or 5 is the same (see Fig. 5.2 for the $I = 4$ fit). Consequently, a discrimination between these two spin values can not be made relying on the hyperfine spectra only. However, a conclusion can be drawn from the discrepancy between the experimental and calculated (using the additivity rule (equation (2.33))) magnetic moments. The following magnetic moments are obtained from the fits of the hyperfine spectra: $\mu(^{180}\text{Tl}, I = 4) = -0.58(2)$ n.m. and $\mu(^{180}\text{Tl}, I = 5) = -0.59(2)$ n.m., assuming spin $I = 4$ and $I = 5$ respectively.

Using the additivity relation (equation (2.33)) to estimate the magnetic moments leads to $\mu(^{180}\text{Tl}, I = 4, [\pi 3s_{1/2} \otimes \nu 1h_{9/2}]) = -0.61$ n.m. and $\mu(^{180}\text{Tl}, I = 5, \pi 3s_{1/2} \otimes \nu 1h_{9/2}) = +2.3$ n.m.

The value used in the additivity rule calculations was, for the $\pi 3s_{1/2}$ configuration, a magnetic moment $\mu(\pi 3s_{1/2}) = 1.6$ n.m. from the neighboring odd thallium isotopes. Unfortunately there are no experimental data available involving the $\nu 1h_{9/2}$ orbital, so a shell model estimation [117, 118] was used. With $g_s = 0.6$ $g_{s,free}$ and $g_l = 0.05$ (see [90]), $\mu(\nu 1h_{9/2}) = 0.69$ n.m. is obtained. Other reasonable choices for g_s and g_l give for $I = 4$ $\mu(^{180}\text{Tl}, I = 4, [\pi 3s_{1/2} \otimes \nu 1h_{9/2}]) = -0.3 \dots -0.6$ whereas for $I = 5$ $\mu(^{180}\text{Tl}, I = 5,$

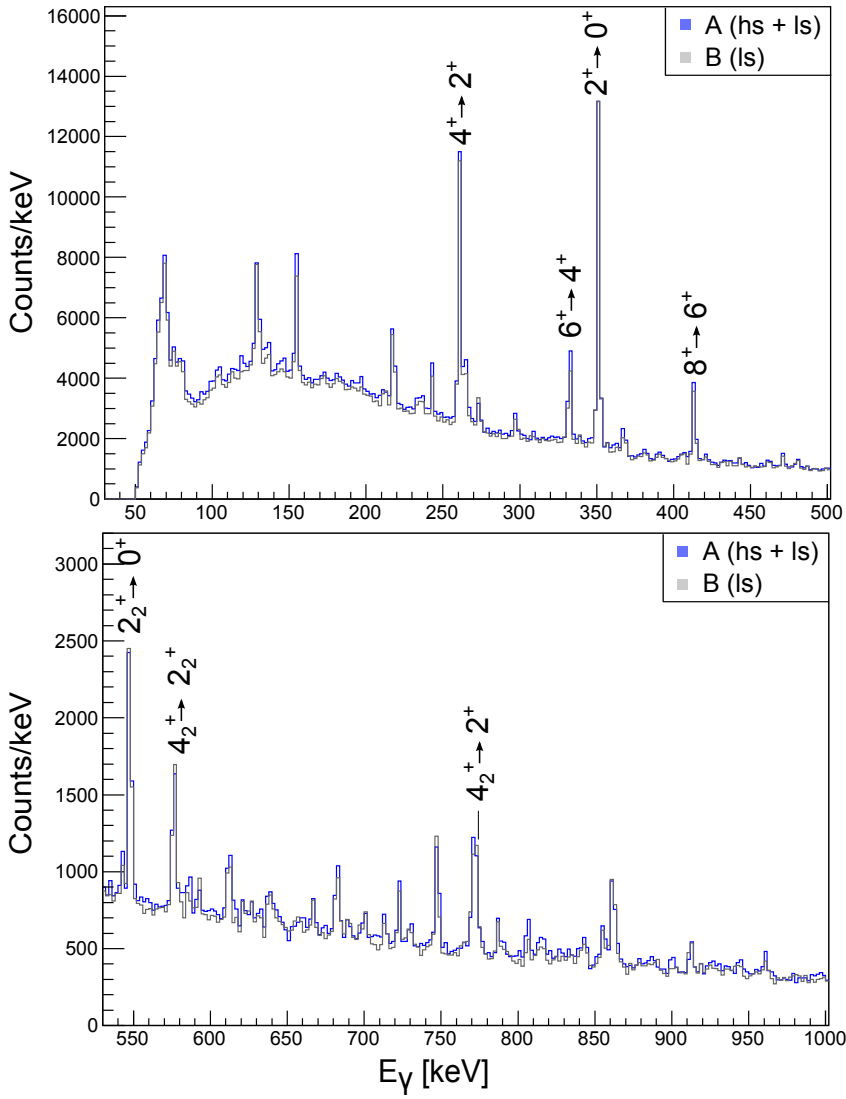


Figure 5.16: [Color] γ spectra for the frequency range A (blue) and B (grey) collected at mass 182. A number of transitions in ^{182}Hg , fed in the β decay of ^{182}Tl , are indicated.

$[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]) = +2 \dots +2.4[90]$. So a $I = 5$ spin assignment leads to a large, positive magnetic moment, where a $I = 4$ spin assignment gives a comparatively small and negative magnetic moment. Moreover, other possible (and less probable) configurations for spin $I = 4$ or 5 all give a large positive magnetic moment.

Based on the above arguments, we can thus propose a $I = 4$ spin assignment for the ground state of ^{180}Tl with configuration $[\pi 3s_{1/2} \otimes \nu 1h_{9/2}]$. As mentioned in the previous section, it should be noted that the HFS shape and the magnetic moment of low-spin state in ^{182}Tl ($-0.53(3)\text{n.m.}$) obtained with the $I = 4$ assumption point to the same configuration assignment for this state.

5.5 Mean-square charge radii of $^{179-184}\text{Tl}$

The mean-square charge radii $\delta\langle r^2 \rangle$ for the different states in $^{179-184}\text{Tl}$ deduced in this work are given in Table 5.1 and have been determined using equation (5.1). The isotope shift and charge radii given in Table 5.1 are relative to the values of stable ^{205}Tl , measured during the same experimental campaign.

Figure 5.17 shows the isotopic dependence of the changes in the mean-square charge radii $\delta\langle r^2 \rangle$ for the thallium isotopes to get insight into the development of deformation in the isotopic chain. The dashed lines represent the droplet model predictions with different mean-square deformations (see also chapter 2). The new data extend the previously known isomeric chain for both sides ($N > 112$, $N < 106$). These results come from measurements at the ISOLDE and IRIS [7] facilities.

These data show a long chain (N from 102 to 116) of the (intruder) isomeric states with the deformation remarkably greater than the deformation of the ground state. Along with the previously measured $^{186}\text{Tl}^{m2}$ ($I = 10$) [7], a similar large isomer shift was observed also for the intruder state in odd-odd ^{184}Tl .

In Fig. 5.18, 5.19 and 5.20 the relative changes in $\delta\langle r^2 \rangle$ for the even-neutron and odd-neutron thallium, lead, bismuth and mercury isotopes are shown to illustrate the similarities between the different isotopic chains of these elements [119]. For the sake of clarity, the results for the even-neutron and odd-neutron nuclei are presented separately. The values of $\delta\langle r^2 \rangle_{N,126}$ are normalized within each isotopic chain to $\delta\langle r^2 \rangle_{122,124}$ to allow for a comparison between these chains, taking aside the (rather great) uncertainties on the different electronic factors. Data are from Refs. [21, 59, 7, 120, 121] were used.

In the odd-neutron isotopes shown in Fig. 5.18 it is seen that the thallium,

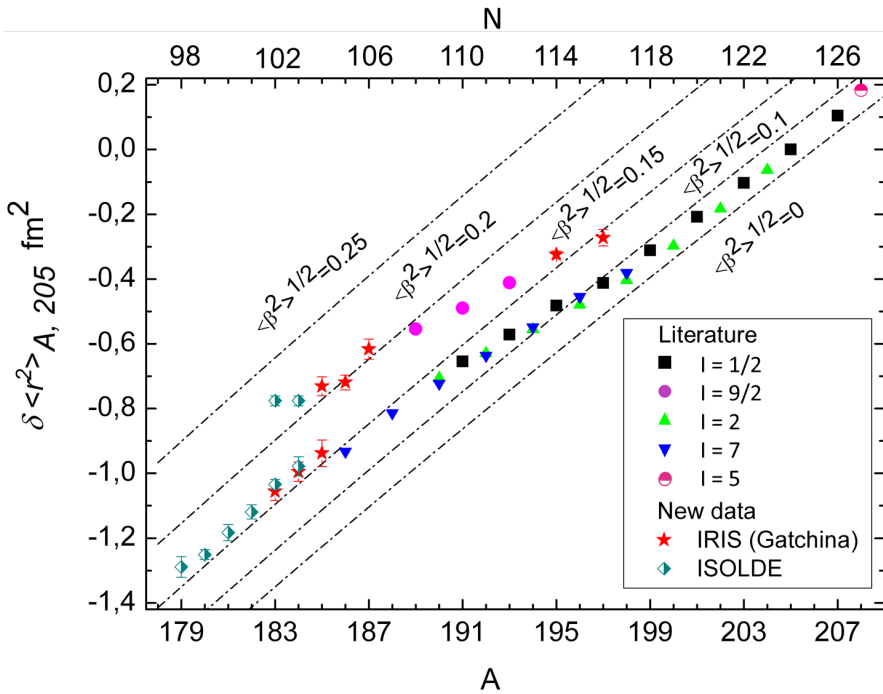


Figure 5.17: [Color] Isotopic dependence of the mean-square charge radii $\delta\langle r^2 \rangle$ for the thallium isotopes. The dashed lines represent the droplet model predictions with different mean-square deformations (see section 2.35).

mercury and bismuth radii trend does not deviate substantially from that of the lead isotopes.

In Fig. 5.19 it is clearly seen that, in contrast to the mercury isotopic chain, no strong odd-even staggering of the charge radii of thallium ground states is observed, and these states preserve their near-spherical shape in the ground states even at neutron midshell and beyond ($A < 186$). A similar smooth behavior of $\delta\langle r^2 \rangle$, which testifies to the persistence of the near-spherical shape, was previously observed for the lead isotopes ($N = 100-126$), whereas the well-known pronounced deviation from this behavior, connected with the onset of permanent prolate deformation, was found for the adjacent mercury ($Z = 80$, [115]) and gold ($Z = 79$, [19]) nuclei. This can also be seen in Fig. 1.4.

In Fig. 5.20 it is seen that the even-neutron thallium and mercury radii follow the pattern of the lead radii even below neutron midshell at $N = 104$.

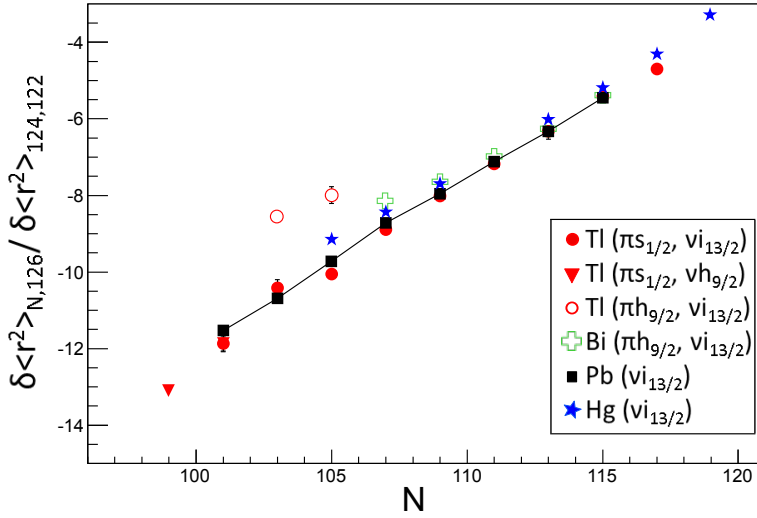


Figure 5.18: [Color] Relative changes in $\delta \langle r^2 \rangle$ for a number of odd-neutron thallium, lead, mercury and bismuth isotopes. Legend is given in the inset.

The bismuth isotopes follow the lead trend until $N = 110$ and deviate from this trend at $N < 110$. For a more detailed description of the deformation change of the bismuth isotopes we refer to [120].

5.5.1 Isomer shift in ^{183}Tl

The excitation energy for the $h_{9/2}$ intruder isomeric states reaches a minimum around $A = 187$ – 189 . In Ref. [22], calculations of deformations and excitation energies for a odd-mass thallium isotopes between $A = 181$ and 199 , using the shell correction method with a Woods-Saxon potential and a monopole pairing residual interaction, predicted that the difference in deformation (i.e., isomer shift) between ground state and isomeric state is nearly constant or even decreases below $A = 187$. However, an unexpected growth of the isomer shift and mean-square deformation relative to heavier odd-mass isotopes has been observed for ^{185}Tl in Ref. [7]. The authors refer to [4] where the mixing and repulsion of two $9/2^-$ configurations situated 299 keV from each other in ^{183}Tl was assumed. With the band built on top of the $h_{9/2}$ proton intruder configuration are two bands associated, a strongly coupled spin sequence and a decoupled spin sequence which are interpreted as weakly deformed oblate

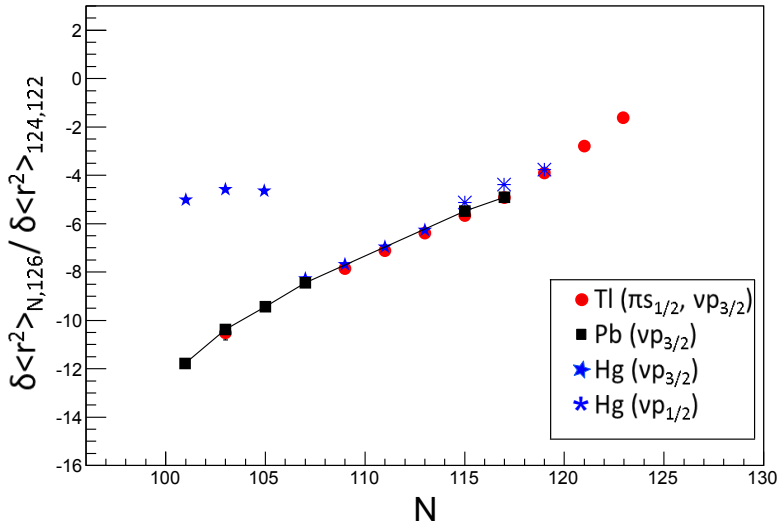


Figure 5.19: [Color] Relative changes in $\delta\langle r^2 \rangle$ for a number of odd-neutron thallium, lead and mercury isotopes. Legend is given in the inset.

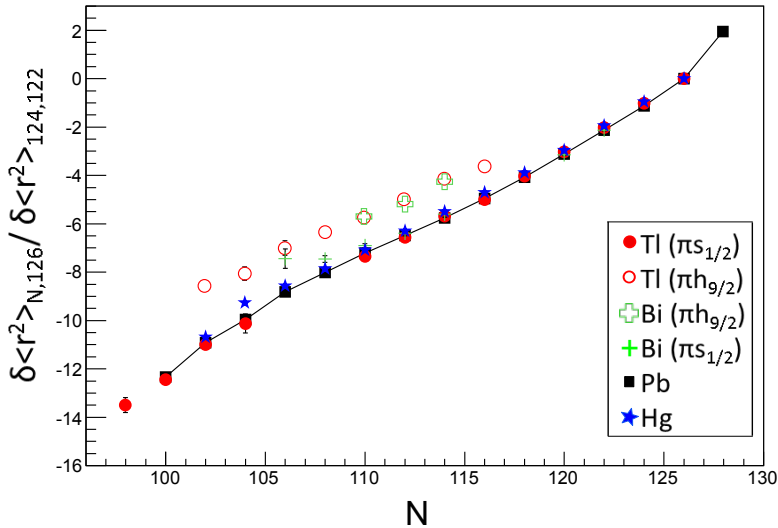


Figure 5.20: [Color] Relative changes in $\delta\langle r^2 \rangle$ for the even-neutron thallium, lead, mercury and bismuth isotopes. Legend is given in the inset.

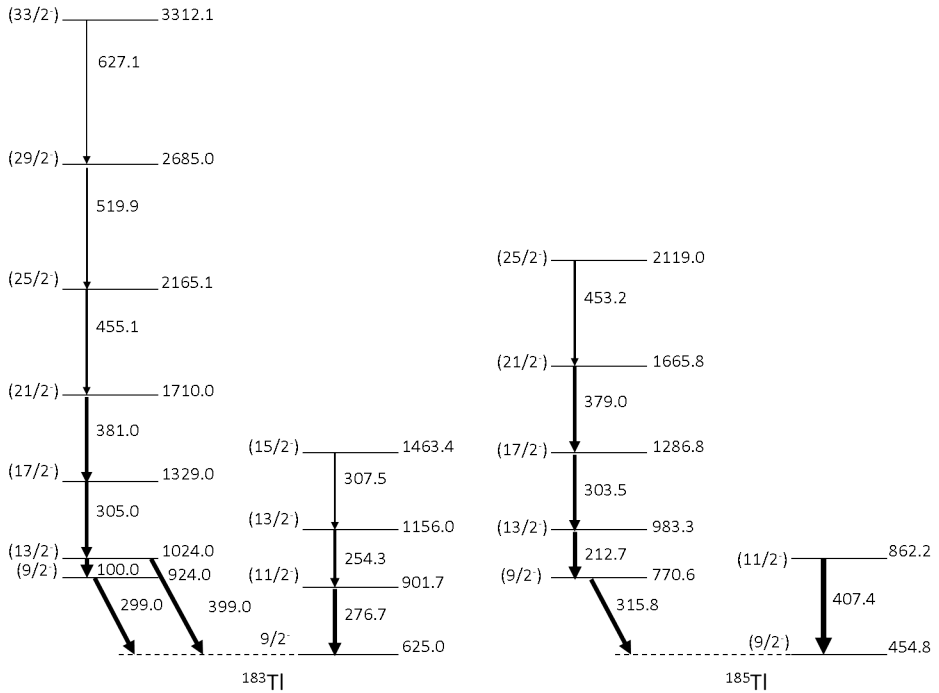


Figure 5.21: Part of the band structure above the $9/2^-$ state in $^{183,185}\text{Tl}$. All energies are in keV. Data are from [8].

and strongly deformed prolate structures [23]. If the band heads of these two configurations are in fact mixing, the mean-square deformation of the $9/2^-$ isomer would increase, compared to the moderate oblate deformation of the heavier thallium isotopes, as the absolute value of the deformation of the prolate deformed $9/2^-$ states is markedly greater than that of the oblate deformed $9/2^-$ states. A similar mixture may be expected in ^{185}Tl as the corresponding $9/2^-$ states are also situated only 316 keV from each other, which might explain the observed increase in isomer shift in ^{185}Tl [7]. Part of the band structure above the $9/2^-$ state in $^{183,185}\text{Tl}$ is given in Fig. 5.21.

The isomer shifts between ground ($1/2^+$) and isomeric (intruder) state ($9/2^-$) in the odd thallium isotopes are shown in Fig. 5.22. The expected increase relative to ^{185}Tl of the isomer shift for ^{183}Tl is indeed observed and supports the increased mixing between the weakly deformed oblate and strongly deformed prolate structures.

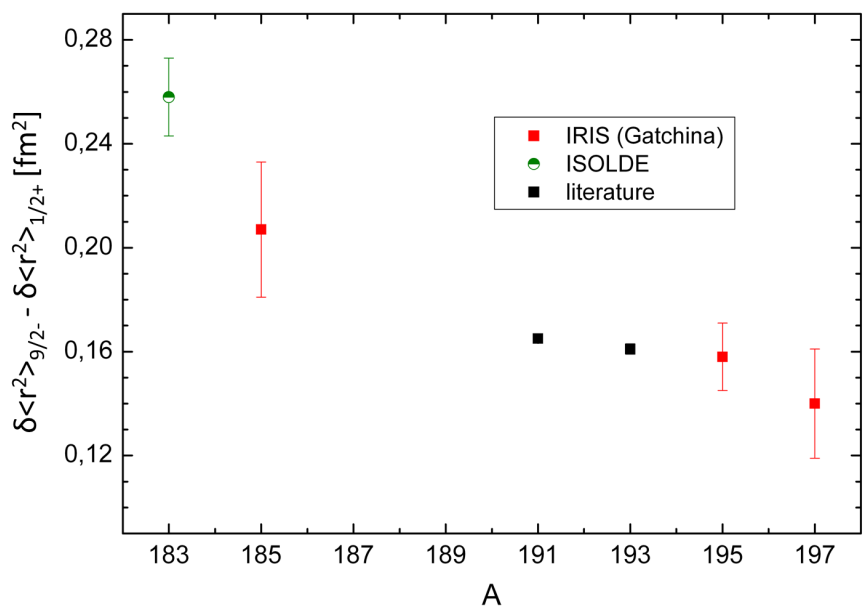


Figure 5.22: [Color] Isomer shift for the $h_{9/2}$ intruder configuration compared to the $s_{1/2}$ ground state in the odd-mass thallium isotopes. Legend is given in the inset.

6 | Conclusions and outlook

6.1 Summary

The neutron-deficient thallium isotopes are situated in the light-lead region of the nuclear chart, where complex nuclear physics properties have been observed such as shape coexistence and intruder states. Shape coexistence has been identified experimentally as well as theoretically in thallium and also other nuclei in this region. Different shapes, whose structure has been linked to specific proton orbitals above and below the $Z = 82$ shell closure, are present at low energy in the neutron-deficient odd-mass thallium nuclei. Next to spherical proton-hole configurations, one particle-two hole (1p-2h) configurations, involving orbitals from above the $Z = 82$ shell closure, that lead to deformation, are identified in the odd as well as odd-odd thallium nuclei. Since thallium has one proton missing in the major proton shell ($Z = 81$), and when approaching neutron mid-shell at $N = 104$, the interpretation of the results is based on the single-particle properties on one hand and on shape coexistence phenomena on the other hand.

However, due to the reciprocity of these different phenomena, the decay and level schemes of e.g. the thallium nuclei studied in this work, are complicated and the spectroscopic interpretation of the underlying configurations of the observed states is often puzzling. Consequently, a combination of different experimental techniques is necessary to unravel the structure of such nuclei. In this work, laser-assisted decay spectroscopy is combined with an optical-spectroscopy study of neutron-deficient thallium isotopes at the ISOLDE facility. This dual approach was essential to identify the different decay branches and disentangle the decay schemes of the different long-lived states in the thallium isotopes. The extracted magnetic moments and mean-square charge radii in the laser spectroscopic study are often an important chain in the interpretation of the decay spectroscopy data.

The IT of the (10^-) isomeric state in ^{184}Tl has been studied, where the precisely

determined excitation energy of this state extends the $(10^-) \rightarrow (7^+)$ energy systematics beyond neutron mid-shell and confirms the parabolic behavior of the excitation energy as a function of neutron number. From the determined half-life value and branching ratios, the retardation factors of the depopulating E3 and M2 transitions were determined and compared with retardation factors in neighboring odd-mass and even-mass thallium isotopes. Combined with the information on the published reduced widths of the α decay of $^{187-192}\text{Bi}$ populating levels in daughter nuclei $^{183-188}\text{Tl}$, a confirmation of the proton 1p-2h intruder character of the (10^-) isomer and an interpretation of the levels in ^{184}Tl fed in the isomeric decay in terms of $[\pi 3s_{1/2} \otimes \nu 1i_{13/2}]$ and $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configurations could be obtained. From the results of the laser spectroscopic study, it was observed that the magnetic moment of the (7_1^+) state shows increased importance of the $[\pi 2d_{3/2} \otimes \nu 1i_{13/2}]$ configuration, compared to respective magnetic moments in the heavier odd-odd thallium isotopes [7, 84] (see also chapter 5). The similarity of the charge radius of the (10^-) isomeric state with the charge radii of the $\pi h_{9/2}$ intruder states in heavier odd and odd-odd thallium isotopes supports the proton intruder character of this state in ^{184}Tl .

Through the observation of hindered and unhindered α decay, low-lying states, possibly intruders, in the daughter nuclei can be selectively studied and identified, yielding direct information on the excitation energy, decay pattern and possible configurations involved. In this work, the α decay of $^{182,184}\text{Tl}$ has been studied. The α decay of the three long-lived states in ^{184}Tl , (10^-) , (7^+) and (2^-) , could be disentangled due to a purification procedure that could be applied to the singles α -decay energy spectra. The previously known and a number of new α -decay lines were assigned to the decay of the (2^-) state and, for the first time, α -decay branches for the (10^-) and (7^+) states were identified. Also for ^{182}Tl new α -decay lines have been identified and a lower limit of the α -decay branching ratio could be determined. Clear evidence was found for the presence of two isomeric states in ^{182}Tl from β -decay studies [65] and the obtained narrow-band laser scans in our study strongly support this observation [84]. All α lines assigned in our study have a hyperfine structure pattern testifying to the predominance of the low-spin state decay in these lines. Out of the narrow-band laser scans, there is evidence for α decay from the high-spin state in the region around 6250 keV, however no further information can be extracted. Detailed α - γ coincidence analysis has been performed for both $^{182,184}\text{Tl}$, however only partial α -decay schemes could be constructed for both isotopes due to the complexity of the α -decay patterns, limited knowledge on the structure of the daughter nuclei and lack of γ - γ coincidences. Reduced α -decay widths and hindrance factors have been calculated for the different α -decay branches in both isotopes and from a comparison with values obtained in neighboring odd-A and even-A thallium isotopes, states with possibly similar or completely different structures

were identified. Only the ^{184}Tl (10^-) state is α decaying unhindered to a state in ^{180}Au , all other α -decay channels observed in this study are hindered, indicating strong structural changes between parent thallium and daughter gold isotopes.

With the aim to extend the previously known isomeric chain beyond neutron mid-shell at $N = 104$, $^{179-184}\text{Tl}$ isotopes have been studied using the in-source laser spectroscopy technique at the ISOLDE facility. The results show that the newly measured magnetic moments follow the isotopic trend for the heavier nuclei with the same spin reasonably well, pointing to the smooth isotopic change in the nuclear structure of the thallium nuclei at neutron mid-shell and beyond. In contrast to the mercury isotopic chain, no strong odd-even staggering of the charge radii of thallium ground states is observed, and these states preserve their near-spherical shape even at neutron mid-shell and beyond ($A < 186$). A similar smooth behavior of the charge radii, which testifies to the persistence of the near-spherical shape, was previously observed for the lead isotopes ($N = 100-126$), while the well-known pronounced deviation from this behavior, connected with the onset of permanent prolate deformation, was found for the adjacent mercury ($Z = 80$, [115]) and gold ($Z = 79$, [19]) nuclei. The in this work measured charge radii of the thallium isomeric states involving the $\pi h_{9/2}$ intruder orbital are significantly larger than those of the ground states, thus following the systematics of the previously measured heavier isotopes.

6.2 Outlook

The detailed α -decay study of $^{182,184}\text{Tl}$ presented in this work revealed the complex structure of daughter gold isotopes. Low-energy, highly-converted transitions, crucial to construct the full level schemes of these odd-odd isotopes, have not been observed in the present study. New experimental data combining a windmill-type implantation chamber with, for example a LeGe detector to detect these low-energy γ transitions and a dedicated detection system for electrons, and the combination of other experimental techniques, such as in-beam γ -ray spectroscopy and laser spectroscopy studies, with our decay data would contribute to establish the decay schemes of these isotopes. For example, as pointed out in chapter 4, the combination of low-lying states in ^{178}Au identified in this work through α - γ coincidences, the laser spectroscopy studies at ISOLDE and the data obtained at a recently performed in-beam study of ^{178}Au at RITU should help to establish the full level scheme of ^{178}Au . A high-spin and low-spin α -decaying state have already been identified in the latest laser spectroscopy studies at ISOLDE [111].

Coulomb excitation studies of $^{182-188}\text{Hg}$ have been performed [122] with 2.85 MeV per nucleon mercury beams from REX-ISOLDE. Using higher beam energies at HIE-ISOLDE where post-accelerated beams with energies up to 10 MeV per nucleon will be produced [123, 124], will result in higher cross sections for multi-step Coulomb excitation, providing more information about e.g. reduced E2 transition probabilities, quadrupole moments and ground state deformations in these isotopes. For these experiments the electron detector SPEDE, which is constructed jointly by the universities of Liverpool and Jyväskylä, will provide a direct way of detecting E0 transitions [125]. This might improve the uncertainty on the observed E0 decay of the populated states.

A Coulomb excitation experiment of $^{182,184}\text{Hg}$ has been proposed [126] using the higher beam energies of HIE-ISOLDE. However, crucial for the analysis of the Coulomb excitation data is the knowledge of the conversion coefficients and branching ratios of the lowest lying 0^+ , 2^+ , 4^+ states, determined in the β -decay study of $^{182,184}\text{Tl}$. In our latest β -decay study of these isotopes, the detection set-up was not optimal and the results are influenced by e.g. the efficiency of the HPGe detectors which is strongly affected by the high γ -multiplicity in the β decay. It is thus highly desirable to study the β decay of $^{180,188}\text{Tl}$ with an optimized experimental setup to obtain reliable input data for the Coulomb excitation study. The recently developed ISOLDE Decay Station (IDS) is suitable for this purpose. At the IDS, a dedicated γ -ray spectroscopy setup is currently installed using Clover HPGe detectors and a compact silicon and plastic based setup is foreseen in the near future for e.g. high-resolution electron spectroscopy studies.

Despite the wealth of experimental data available for the doubly magic nucleus ^{208}Pb ($Z = 82$, $N = 126$) and its closest neighbors, the current body of experimental information regarding the more neutron-rich quadrant, defined by $Z \leq 82$ and $N > 126$, is still scarce, while there are already indications of unexplained features of the nuclear structure in the region (see e.g. [127]). The reason for the limited spectroscopic information lies in the experimental difficulties to access this region of the nuclear chart. The synthesis of neutron-rich trans-lead nuclei in fusion-evaporation reactions is experimentally problematic due to the strong competition with fission; on the other hand, spallation reactions suffer from high contamination levels from more abundantly produced isobars. However over the years other techniques have been used to explore this region, e.g. fragmentation reactions [128, 129] and multi-nucleon transfer reactions on ^{238}U target [130, 131]. At ISOLDE, the combination of the RILIS and the pulsed release method has paved the way to reach these radioactive species by efficiently suppressing the isobaric contamination [132] which lead to the study of e.g. $^{215-218}\text{Bi}$ and ^{215}Pb and the newly developed Laser Ion Source Trap (LIST) target has given access to study the decay properties of ^{219}Po . These

experimental techniques certainly open the ground to study more neutron-rich isotopes in this region in the near future.

Currently the mean-square charge radii of $^{187,189}\text{Tl}$ ground states have not been determined due to the much higher production of the (intruder) isomeric state relative to that of the ground state and the absence of pure $J = 1/2$ γ lines to monitor the hyperfine structure pattern of the ground state. However, these charge radii are of considerable importance to get insight in the development of the increase in the intruder isomer shift (see Fig. 5.22). A possibility to determine these ground-state charge radii is by using ISOLTRAP's multi-reflection time-of-flight mass separator/spectrometer (MR ToF MS) [133]. The energy difference between the (intruder) isomeric state and the ground state is in both $^{187,189}\text{Tl}$ about 300 keV and these states could thus possibly be separated by MR ToF MS. The applicability of the MR ToF MS for scanning the hyperfine structure and separating isomeric states has already been proven successful in a number of gold, astatine and mercury isotopes during recent experimental campaigns. However, the intruder isomeric states in $^{188,189}\text{Tl}$ will be accessible experimentally by combining RILIS with the IDS.

Continuous efforts are made to improve the in-source resonance ionization spectroscopy technique. For example, the introduction of Doppler-free methods would lead to high-precision experiments, giving access to e.g. more accurate values of the hyperfine anomaly in the case of gold or bismuth isotopes and a better determination of the quadrupole moments from the hyperfine spectra.

In laser spectroscopy experiments spectral linewidths are required to be as close as possible to the intrinsic natural linewidths of the atomic transitions of interest. For the successful study of atomic properties of elements with particularly small hyperfine splitting or high sensitivity to atomic collisions, a novel approach such as in-gas-jet laser spectroscopy would be the technique of choice.

Radioactive Ion Beam facilities such as ISOLDE at CERN, SPIRAL1 at GANIL and TRIUMF currently push the limits of spectroscopy with exotic nuclei. However, these facilities rely on the Isotope Separator On-Line (ISOL) method using high temperature target ion source systems, which can be chemically restrictive [61]. For example, beams of short-lived isotopes from chemical elements that have refractory properties have very low intensity or are not produced at all. In addition, the production mechanism used at these facilities does not allow to reach the heavy element region. To overcome the present limitation a novel laser spectroscopy technique, is being developed. The so-called Heavy Element Laser Ionization Spectroscopy (HELIOS) project at KU Leuven expects to address the issues that are currently limiting the experiments. The technique will be fast, selective and very sensitive [134]. Using the very intense ion beams provided by SPIRAL2 at GANIL, it will allow for the study

heavy elements using laser spectroscopy. A generic In Gas Laser Ionization and Spectroscopy (IGLIS) setup will be commissioned and tested at the HELIOS laboratory in Leuven and after a test and optimization period, the setup will be installed at the Super Separator Spectrometer (S3) in GANIL, where full operation in on-line conditions is intended.

A | Experimental determination of efficiency for Si detectors

In this appendix different methods are described to determine the absolute or relative detection efficiency of the Si detectors in the Windmill (WM) detection setup using mainly the ^{241}Am calibration sources. Data from several ISOLDE experiments are discussed: IS466-III (May 2011) , IS534 (May2012) and IS534-IV (May2015). The data presented in this appendix have been analysed by Lars Ghys and Simon Sels. Thanks guys!

In Fig. A.1, a schematic representation of the WM detection system is given. The position of the four Si and two Ge detectors is indicated with respect to the incoming beam. This configuration is the same for all experiments discussed in this appendix. During the experiments a collimator with a diameter of 5 mm was used. The maximum diameter of the implanted ion beam in the carbon foil can thus be considered 5 mm. From the technical drawings of the WM system, the distance between the different detectors and the source can be deduced. The typical distance of the Si1 detector from the source is about 7 mm, while for Si2 this is about 4 mm. The distances lead to solid angles close to 20 % for Si1 and 30 % for Si2. The distance of the Si3 detector from the source is about 4 mm, since it is positioned in an identical manner as Si2. For Si4, the distance from the source is difficult to determine since this detector is mounted on a rail and its position is thus not fixed. Based on the geometry, a distance between 4 and 7 mm is a reasonable assumption. For the Si3 and Si4 detectors, these distances lead to solid angles close to 30 % for Si3 and around 25-30 % for Si4.

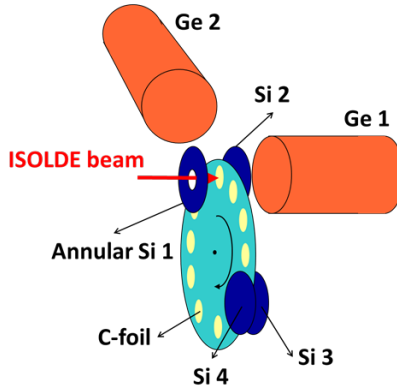


Figure A.1: Schematic representation of the WM detection system. The position of the four Si and two Ge detectors is indicated with respect to the incoming beam.

A.1 ^{241}Am calibration sources

The discussion in this text is largely based on measurements with two weak ^{241}Am calibration sources mounted on the wheel of the WM. One of the sources was installed downstream and the other upstream. In other words, one of the sources faces either Si1 or Si4, and the other faces either Si2 or Si3. Both sources, with serial numbers NX532 and NX533 were manufactured by Eckert & Ziegler and are of the type AMR11. The nominal activity in April 2011 was around 50 Bq, but with a rather large uncertainty of about 30%. The ^{241}Am nuclei are deposited on a stainless steel disc, 25 mm in diameter, 0.5 mm thickness. The active area of the sources has a diameter of 7 mm. The most intense α -decay of ^{241}Am has an energy of 5485 keV and is coincident with a 60 keV γ line.

A.2 Run IS466-III – May 2011

This section discusses the Si detection efficiency in the IS466-III run, from which the measurements were performed in May 2011 at ISOLDE. The main purpose of this experiment was the experimental investigation of β -delayed fission in ^{202}Fr . The detector properties are given in Table A.1.

Table A.1: Overview of the different detectors used during IS466-III.

Detector	Type	Serial Number
Si1	SB ^a , Ortec	43-051/F ^c
Si2	SB ^a , Ortec	47-038D
Si3	PIPS ^b , Canberra	74256
Si4	PIPS ^b , Canberra	74257
Ge1	HPGe, Canberra	GC9019, b90008
Ge2	HPGe, Canberra	GC7023, b88045

^a Surface Barrier (SB)

^b Passivated Implanted Planar Silicon (PIPS)

^c Annular, hole of 8 mm

Table A.2: Absolute efficiency of Si1 and Si2 for IS466-III, determined by α - γ coincidences.

File name	Detector	N_γ	$N_{\alpha\gamma}$	ϵ_{Si} (%)
test0013	Si1	1337(49)	121(11)	9(1)
test0012	Si2	2370(61)	352(31)	15(1)

Before the run, calibration data were taken using the two ^{241}Am sources (~ 50 Bq) mounted in the wheel. Since the Ge detectors were also installed, the absolute efficiency can be determined for the Si1 and Si2 detectors, using α - γ coincidences between the 5485 keV ^{241}Am α decay and 60 keV γ ray. The absolute efficiency ϵ_{Si} is found as the ratio between the number of α - γ coincidences $N_{\alpha\gamma}$ between Ge1 and Si1 or Si2 and the number of singles γ detected in Ge1 N_γ . The results are summarized in Table A.2. This method cannot be used for the detectors Si3 and Si4 as the position of these detectors is too far from the Ge detectors.

The activity of the sources was also calculated, using the absolute efficiencies of Si1 and Si2 determined above (see Table A.2). The results are given in Table A.3. The calculated activity is consistent with the activity of 50 Bq, which was requested for these sources. According to the source specifications, the uncertainty on the source activity may be up to 30%.

The relative efficiencies Si2/Si3 and Si1/Si4 can be deduced by comparing recorded α rates in the different detectors from the ^{241}Am sources. Using the deduced absolute efficiencies for Si1 and Si2 from α - γ coincidences in Table A.2, also the efficiencies for Si3 and Si4 can then be determined. The results are

Table A.3: Absolute efficiency of all four Si detectors for IS466-III, determined by α - γ coincidences, number of α /s and calculated activity of the ^{241}Am source.

File name	Detector	α /s	ϵ_{Si} (%)	Activity (Bq)
test0013	Si1	4.46(1)	9(1)	$\rightarrow$ 50(6)
test0012	Si2	8.35(1)	15(1)	$\rightarrow$ 56(4)
test0014	Si3	11.80(2)	21(2)	$\leftarrow$ 56(4) ^a
test0012	Si4	8.16(1)	16(2)	$\leftarrow$ 50(6) ^b

^a Measurement with the same calibration source as Si2

^b Measurement with the same calibration source as Si1

Table A.4: Number of detected α particles and corresponding source for the four Si detectors.

Detector	N_α	Source
Si1	$2.579(3) \times 10^6$	^{202}Fr
Si2	$4.781(4) \times 10^6$	^{202}Fr
Si3	$2.700(2) \times 10^5$	^{198g}At
Si4	$1.690(2) \times 10^5$	^{198g}At

summarized in Table A.3. Since the pulser was not properly installed at this point, the measurement time was found by the difference between the timestamp of the first and last recorded event in the Si detectors, thereby ignoring possible dead-time effects. (note: the used calibration sources are rather weak, thus dead-time effects are expected to be minimal).

Finally, the relative efficiency of Si1 to Si2 and Si 3 to Si 4 can also be deduced from implanted activity on the carbon foil during the experiment. Table A.4 gives the number of detected α particles N_α in Si1,2,3,4 from the nuclei ^{202}Fr (g.s. $T_{1/2} = 0.30(5)$ s, i.s. $T_{1/2} = 0.29(5)$ s) and ^{198g}At ($T_{1/2} = 3.8(4)$ s) [8].

The efficiency ratio $\epsilon_{Si1}/\epsilon_{Si2}$ and $\epsilon_{Si3}/\epsilon_{Si4}$ can be deduced from the data in Table A.4, as well as from the ^{241}Am source. The ratios extracted using both methods are compared in Table A.5.

The values in this table are, within error bars, reasonably consistent. The dimensions and intensity distribution of the implanted sources are most probably different to the dimensions of the calibration sources which might influence the results.

Table A.5: Comparison of the efficiency ratio using implanted activity and ^{241}Am sources.

Detectors	Implanted activity	^{241}Am
$\epsilon_{Si1}/\epsilon_{Si2}$	0.539(1)	0.6(1)
$\epsilon_{Si3}/\epsilon_{Si4}$	1.60(2)	1.3(2)

A.3 Run IS534– May 2012

In May 2012, a run dedicated to the β -delayed fission of $^{194,196}\text{At}$ and ^{200}Fr was performed. The same detector configuration as in run IS466-III was used here (see Table A.1). Calibration data were taken using the ^{241}Am sources before this run, but the Ge detectors were not connected at the time of these measurements. Consequently, the absolute detection efficiency could not be determined using α - γ coincidences. The pulser was also absent at that time, so the total measurement time was again deduced from the time difference between first and last recorded event.

The number of α decays per second from ^{241}Am recorded for the different detectors are listed in Table A.6. Using the activity of the ^{241}Am sources from Table A.3, the efficiency of the Si detectors could be deduced. It should be noted that Si4 is mounted on a rail, so the distance foil-to-detector may vary from one experiment to the other, which might explain the slight mismatch in calculated efficiency.

The efficiency ratio $\epsilon_{Si1}/\epsilon_{Si2}$ and $\epsilon_{Si3}/\epsilon_{Si4}$ can be deduced from the data in Table A.6. The ratios $\epsilon_{Si1}/\epsilon_{Si2} = 0.6(1)$ and $\epsilon_{Si3}/\epsilon_{Si4} = 1.1(2)$ agree within error bars with those obtained in IS466-III (Table A.5).

Table A.6: Number of α /s, activity of the ^{241}Am source and deduced efficiency for IS534.

File name	Detector	α /s	Activity (Bq)	ϵ_{Si} (%)
testsi1_si30001 ^a	Si1	4.43(1)	50(6)	9(1)
test_si20003 ^b	Si2	8.70(1)	56(4)	16(1)
testsi1_si30001 ^b	Si3	11.90(2)	56(4)	21(2)
test_si40004 ^a	Si4	9.24(1)	50(6)	18(2)

^a Measurement with the same calibration source
^b Measurement with the same calibration source

A.4 Run IS534-IV – May 2015

Experiment IS534-IV at ISOLDE was mainly dedicated to laser spectroscopy of gold isotopes. The detector configuration is shown in Table A.7.

Table A.7: Overview of the different detectors used during IS534-IV. Information on the corresponding position of each detector can be found in chapter 3.

Detector	Type	Serial Number
Si1	SB ^a , Ortec	43-082H ^c
Si2	SB ^a , Ortec	49-186B
Si3	PIPS ^b , Canberra	74256
Si4	PIPS ^b , Canberra	74257
Ge1	LeGe, Canberra	b92523B
Ge2	HPGe, Canberra	GC7023, b88045

^a Surface Barrier (SB)

^b Passivated Implanted Planar Silicon (PIPS)

^c Annular, hole of 6 mm

During a number of calibration measurements on ²⁴¹Am, the Ge detectors as well as a 100 Hz pulser were connected. However, contamination from a previous run (mercury laser spectroscopy) dominated the γ spectra and an absolute efficiency could not be determined from these measurements. No calibration measurements with ²⁴¹Am on detector Si4 were found. The available data is listed in Table A.8. The number of detected α /s is here slightly higher than in the previous runs (IS466-III and IS534, see Tables A.3 and A.6). The efficiency of the different detectors was determined using the activity of the ²⁴¹Am sources. The resulting values are slightly higher than those obtained in the IS466-III and IS534 runs.

Table A.8: Number of α /s, activity of the ²⁴¹Am source and deduced efficiency for IS534-IV.

File name	Detector	α /s	Activity (Bq)	ϵ_{Si} (%)
241Am_Si1_0391	Si1	6.14(1)	50(6)	12(1)
241Am_Si2_0392	Si2	10.90(2)	56(4)	19(1)
241Am_Si1_0391	Si3	12.70(2)	56(4)	23(2)

Table A.9: Number of α particles and corresponding source for IS534-IV.

Detector	N_α	Source
Si1	$1.72(1) \times 10^4$	^{178}Au
Si2	$2.56(2) \times 10^4$	^{178}Au
Si3	218(15)	^{178}Pt
Si4	164(13)	^{178}Pt

The efficiency ratios $\epsilon_{Si1}/\epsilon_{Si2}$ and $\epsilon_{Si3}/\epsilon_{Si4}$ can be determined using implanted activity during the run, see Table A.9. In this case, ^{178}Au was implanted and the α particles from ^{178}Au and its β -decay daughter ^{178}Pt were used.

Using the data from the implanted sources in Table A.9 gives $\epsilon_{Si1}/\epsilon_{Si2} = 0.67(1)$ and $\epsilon_{Si3}/\epsilon_{Si4} = 1.3(1)$. The ratio $\epsilon_{Si1}/\epsilon_{Si2}$ can also be determined using the data from the ^{241}Am calibration source (Table A.8) and equals 0.6(1). The ratios for the implanted and calibration sources are thus consistent.

A.5 Conclusion

From this analysis, it seems the relative detection efficiencies of the different detectors (Si1,2,3,4) are consistent. The efficiency of detectors Si1 and Si2 has been simulated by V. Liberati using GEANT4 for different configurations. Figures A.2 and A.3 show the simulated efficiencies of detectors Si1 and Si2 respectively as a function of the distance between source and detector, the spot size and the size of the hole in the annular detector. From the technical drawings of the WM, the distance of Si1 with respect to the foil is about 7 mm, while Si2 is placed at a distance of about 4 mm. This would result in a detection efficiency of ~ 19 or 22% for Si1 with a hole of 8 or 6 mm respectively and $\sim 32\%$ for Si2. The ratio $\epsilon_{Si1}/\epsilon_{Si2}$ is thus around 0.59 or 0.68 (depending on the size of the hole), consistent with the experimental observations (see Tables A.5 and A.9)

However, the absolute detection efficiency of Si1 and Si2 as deduced in Table A.2 is a factor of two lower than what would be expected from geometrical considerations.

In November 2015 a dedicated off-line measurement was carried out to find the cause of this inconsistency. These measurements showed that the positioning of the detectors can easily vary from experiment to experiment, changing position by as little as 1 mm can have a large impact on the efficiencies of the

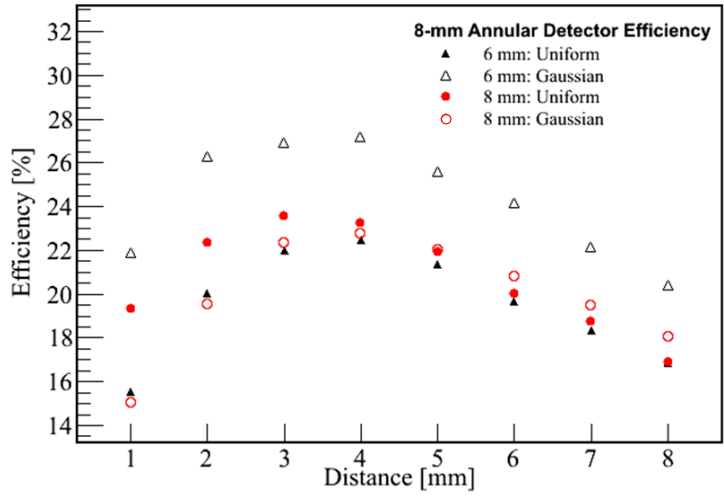


Figure A.2: Simulated efficiency of Si1 using GEANT4.

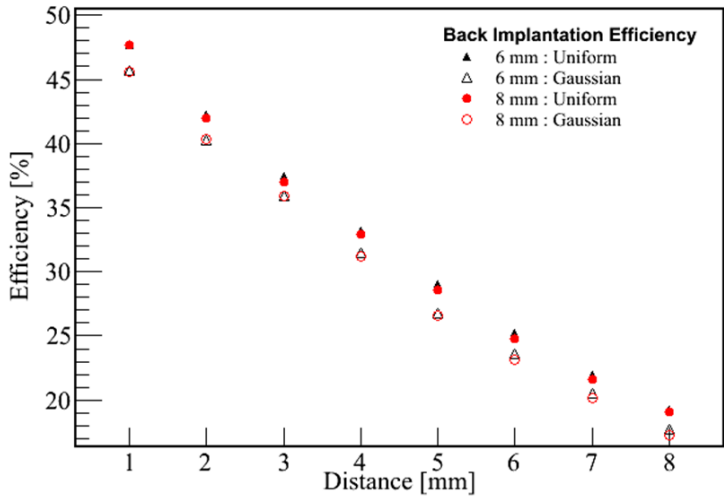


Figure A.3: Simulated efficiency of Si2 using GEANT4.

Table A.10: Deduced efficiency of the four Si detectors by a dedicated off-line measurement.

Detector	ϵ_{Si} (%)
Si1	13.3(8)
Si2	14.6(6)
Si3	21.2(18)
Si4	18.7(22)

Table A.11: Deduced source activity by a dedicated off-line measurement.

Source	Activity (Bq)
NX532	49.8(4)
NX533	49.7(3)

system. In the future the actual position of the detectors will be calculated for each experiment using solid angle calculations, Geant simulations or by measuring during set-up/breakdown of the WM. The absolute efficiencies that were determined during the off-line measurement are given in Table A.10. These confirm the values that were deduced in the previous sections of this appendix.

During this measurement, the activity of the two calibration source was also measured to a high accuracy. The results are given in Table A.11.

B | Efficiency of Si2 for electrons

B.1 Detection efficiency for electrons from the 375 keV E0 transition in ^{184}Hg

The detection efficiency of Si2 for electrons is determined using the conversion electrons (CE) from the 375 keV E0 transition ($0_2^+ \rightarrow 0_1^+$) in ^{184}Hg , where the 0_2^+ is populated in the β^+ /EC decay of ^{184}Tl . The 375 keV E0 transition gives rise to K- and L-CE with energies of 292 and 360 keV respectively. Due to the limited resolution of the Si2 detector, M(and higher order)-CE can not be resolved from L-CE.

Figure B.1 is the electron spectrum in coincidence with the 608 keV γ line. The latter is the $2_3^+ \rightarrow 0_2^+$ (see Fig. B.2). This spectrum is produced looking to coincidences between Si2 and Ge1, within the SI data set and looking to events from 200 ms after each proton pulse (PP) until the next PP in order to avoid events from the (10^-) isomer in ^{184}Tl with a 47.1(7) ms half-life.

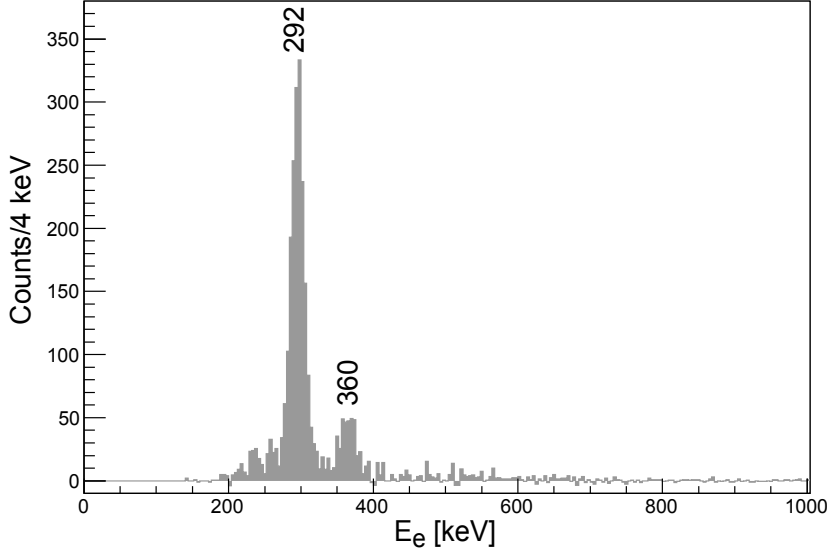


Figure B.1: Electron spectrum in coincidence with the 608 keV γ line. Coincidences between Si2 and Ge1 are shown, using the SI data set and looking to events from 200 ms after each PP until the next PP.

The intensity ratio of the 292 keV K-electrons to the 360 keV L-electrons $I_{Ke}^{292}/I_{Le}^{360}$ in Fig. B.1 is 6.22(66). The ratio of electronic factor $\Omega_{Ke}^{292}(E0)$ to electronic factor $\Omega_{Le}^{360}(E0)$ is 5.85 [55]. From these two ratios it can be concluded that the efficiency ratio $\epsilon_{Ke}^{292}/\epsilon_{Le}^{360}$ is 1.06(11).

The absolute efficiency for detecting electrons at 292 and 360 keV is given by

$$\epsilon_e^{292} = \frac{I_e^{292}}{I_{\gamma}^{608}} \times \frac{\Omega_{TOT}(E0)}{\Omega_{Ke}^{292}(E0)} = 7.6(16)\% \quad (B.1.1)$$

$$\epsilon_e^{360} = \frac{I_e^{360}}{I_{\gamma}^{608}} \times \frac{\Omega_{TOT}(E0)}{\Omega_{Le}^{360}(E0)} = 7.2(17)\% \quad (B.1.2)$$

where $I_e^{292,360}$ is the intensity of 292,360 keV electrons in the spectrum of Fig. B.1 respectively and I_{γ}^{608} is the number of 608 keV triggers in the singles γ spectrum of Ge1 in the SI data set from 200 ms after the PP until the next PP.

It should be noted that the number of 608 keV triggers is determined using the number of 617 keV triggers in the singles spectrum under the same conditions,

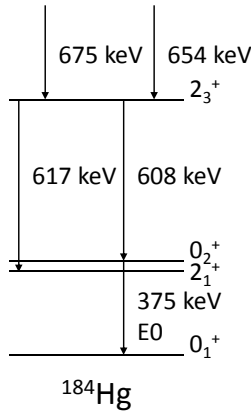


Figure B.2: Partial level scheme of ^{184}Hg , including the 608 and 617 keV γ lines, the feeding 654 and 675 keV γ lines and the 375 keV E0 transition.

normalized to the intensity ratio of the 608 to 617 keV transitions in coincidence with the 654 and 675 keV γ lines. This is necessary because of the difficulties in determining the 608 keV singles intensity due to a doublet structure. A partial level scheme of ^{184}Hg including these transitions is given in Fig. B.2.

B.2 Determination of α_K^{CE} for the 445 keV γ transition in ^{184}Tl

The pulsed structure of the proton beam together with the short implantation time after each proton pulse enables the discrimination between short- and long-living activity as is shown in Fig. 4.2 of chapter 4. Spectrum 4.2a) is a singles γ -ray energy spectrum collected between 10 and 200 ms after each proton pulse and thus contains relatively more short-living than long-living activity. Spectrum 4.2b) was acquired starting from 200 ms after each proton pulse until the next proton pulse. Many γ rays are present in both spectra, mostly originating from the β decay of ^{184}Tl and from longer-lived daughter activities accumulated in the Windmill chamber. Spectrum 4.2c) results from the subtraction of spectra 4.2a) minus 4.2b), after normalization on the 367 keV γ line. The latter is the $2^+ \rightarrow 0^+$ transition in ^{184}Hg . The resulting spectrum 4.2c) contains only short-living activity. The γ rays indicated with

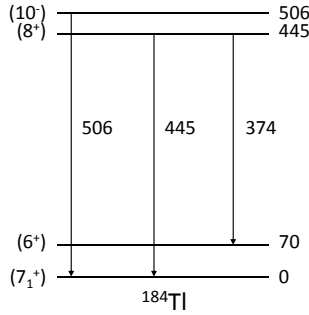


Figure B.3: Partial decay scheme of the (10^-) isomeric state in ^{184}Tl .

their corresponding energy were attributed to the de-excitation of the (10^-) isomeric state.

Using the same procedure, a Si2 electron spectrum is constructed. This spectrum (see Fig. B.4, open spectrum) contains the CE associated with the IT decay of (10^-) state (see Fig. B.3 for a partial decay scheme of the ^{184}Tl IT). The peak at 491 keV corresponds to the L,M-CE from the 506 keV transition. The K-CE from the 506 keV and L,M-CE from the 445 keV transitions give rise to the peak around 421 keV, while the 360 keV peak is attributed to the L,M-CE from the 374 keV and K-CE from the 445 keV transitions. The filled spectrum in Fig. B.4 represents electrons in coincidence with the $K_{\alpha 1}$ line from thallium and is collected between 10 and 200 ms after the proton pulse.

The efficiency of Si2 at 360 keV ϵ_e^{360} (see section B.1) can now be used to calculate the $\alpha_{K(445)}^{CE}$ for the 445 keV transition:

$$\alpha_{K(445)}^{CE} = \frac{I_e^{360}}{\epsilon_e^{360} \times \epsilon_{K_{\alpha 1}} \times \omega_{K_{\alpha 1}}} \bigg/ \frac{I_{\gamma}^{445}}{\epsilon_{\gamma}^{445}} \quad (\text{B.2.1})$$

where I_e^{360} is the intensity of the 360 keV peak in the filled spectrum of Fig. B.4 (thus only due to K-conversion of the 445 keV γ line), $\epsilon_{\gamma}^{445, K_{\alpha 1}}$ is the efficiency in Ge1 for the 445 keV and $K_{\alpha 1}$ lines respectively, I_{γ}^{445} the intensity in the purified (10^-) singles γ spectrum of Fig. 4.2 in chapter 4 and $\omega_{K_{\alpha 1}}$ the fluorescence of thallium $K_{\alpha 1}$ X-rays.

From $\alpha_{K(445)}^{CE} = 0.09(2)$, the mixture M1/E2 of the 445 keV transition can be deduced: $[78(^{+22}_{-26})\% \text{ M1} + 22(^{+26}_{-22})\% \text{ E2}]$ [55].

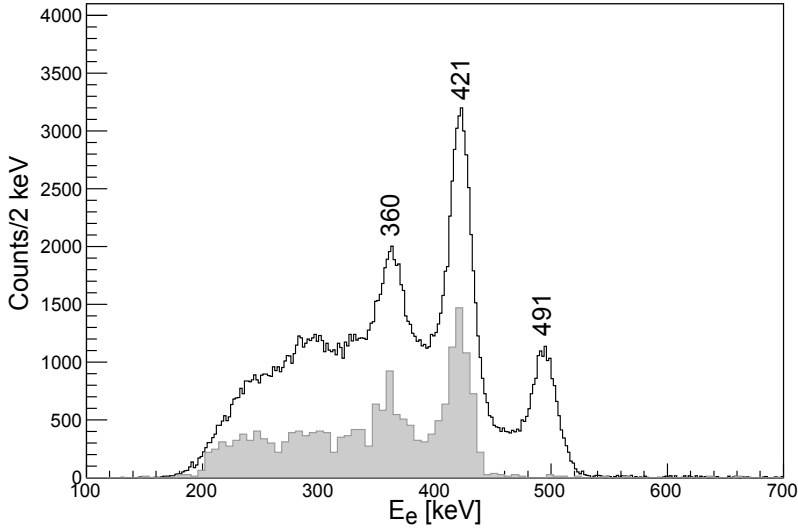


Figure B.4: Electron spectra containing mainly IT activity. The open spectrum is obtained using the purifying procedure for the ^{184}Tl (10^-) state explained in chapter 4. The filled spectrum contains only K-CE from the decay of the (10^-) state by applying a coincidence condition on the thallium K_{α_1} X-rays. It is scaled with an arbitrary factor for visualisation purposes. Electron energies are given in keV.

B.3 Detection efficiency for electrons from the 506 keV E3 transition in ^{184}Tl

The 506 keV transition in ^{184}Tl gives rise to L,M-CE with an energy of 491 keV (see open spectrum of Fig. B.4). Assuming this transition has pure E3 multipolarity, the efficiency for electrons at 491 keV can be determined in the following way

$$\epsilon_e^{491} = \frac{I_e^{491}}{\alpha_{L,M(506)}^{CE}} \times \frac{\epsilon_\gamma^{506}}{I_\gamma^{506}} = 6.4(12)\% \quad (\text{B.3.1})$$

where I_e^{491} is the intensity of the 491 keV peak in the open spectrum of Fig. B.4, $\alpha_{L,M(506)}^{CE}$ the theoretical conversion coefficient for the 506 keV line (assuming pure E3 multipolarity)[55], ϵ_γ^{506} the efficiency in Ge1 for the 506 keV γ line and I_γ^{506} the intensity in the purified (10^-) singles γ spectrum of Fig. 4.2 in chapter

4.

Performing the same calculation for the 421 keV peak in the open spectrum of Fig. B.4, composed of the K-CE from the 506 keV and L,M-CE from the 445 keV transitions, leads to

$$\epsilon_e^{421} = \frac{I_e^{421}}{(I_\gamma^{506}/\epsilon_\gamma^{506} \times \alpha_{K(506)}^{CE}) + (I_\gamma^{445}/\epsilon_\gamma^{445} \times \alpha_{L,M(445)}^{CE})} = 9.5(15) \quad (\text{B.3.2})$$

However, in the filled spectrum of Fig. B.4, the 421 keV peak is composed only of K-CE from the 506 keV. Assuming again E3 multipolarity, this leads to the following efficiency

$$\epsilon_e^{421} = \frac{I_e^{421}}{\alpha_{K(506)}^{CE}} \times \frac{\epsilon_\gamma^{506}}{I_\gamma^{506}} = 9.6(15)\% \quad (\text{B.3.3})$$

where I_e^{421} is the intensity of the 421 keV peak in the filled spectrum of Fig. B.4, $\alpha_{K(506)}^{CE}$ the theoretical conversion coefficient for the 506 keV line (assuming pure E3 multipolarity)[55], ϵ_γ^{506} the efficiency in Ge1 for the 506 keV γ line and I_γ^{506} the intensity in the purified (10^-) singles γ spectrum of Fig. 4.2 in chapter 4. The efficiency for the 421 keV CE using the open or filled spectrum of Fig. B.4 leads to the same value.

B.4 Overview of the results

In Fig. B.5 an overview of the efficiency of Si2 for electrons calculated in the previous sections is given. The efficiency at 421 keV seems to deviate from the other values, however the error bars are large. Still, this value is not used for any further analysis of the thallium decay data.

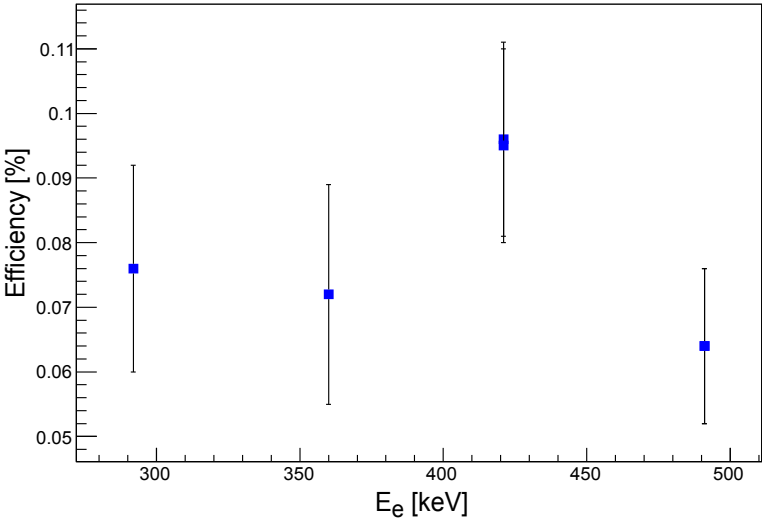


Figure B.5: Summary of the obtained Si2 detection efficiency and corresponding error for electrons.

C | Efficiency of the HPGe detectors using γ - γ coincidences

In the experiments where the data discussed in this work were collected, two single crystal high-purity germanium (HPGe) detectors were placed outside the Windmill chamber to detect γ rays. A detailed description of these detectors is given in chapter 3. Energy and efficiency calibrations were performed using standard calibrated sources of ^{133}Ba , ^{137}Cs , ^{60}Co , ^{241}Am and ^{152}Eu [66]. However, due to the combination of a high γ -multiplicity in the β decay of ^{182}Tl and ^{184}Tl and close-detector configuration, the effective efficiency for the HPGe detectors was lower than that determined by the efficiency calibration using standard calibrated sources. To account for this effect, the efficiency for a number of γ lines, has been determined by γ - γ coincidences.

It should be noted that the (absolute) efficiencies determined using γ - γ coincidences were only used in this work to determine the α -decay branching ratio of the (7^+) state in ^{184}Tl , using the intensity of the 367 keV $2^+ \rightarrow 0^+$ transition in ^{184}Hg and a lower limit of the α -decay branching ratio of the low-spin state in ^{182}Tl , using the intensity of the 351 keV $2^+ \rightarrow 0^+$ transition in ^{182}Hg (see section 4.3 in chapter 4 for a detailed description). In the IT of the ^{184}Tl (10^-) state, the efficiencies determined using the standard calibrated sources were used, since these γ transitions don't suffer from the high-multiplicity effect. In the analysis of the β^+/EC decay of E. Rapisarda [65], only relative efficiencies were used, based on the efficiencies determined by the standard calibrated sources.

C.1 Determination of efficiency using γ - γ coincidences

The efficiency to detect a γ line with a certain energy using γ - γ coincidences is given by the following equation:

$$\epsilon_{\gamma_1} = \frac{I_{\gamma_1-2,coinc} \times (1 + \alpha_{CE}(\gamma_1))}{I_{\gamma_2,singles}} \times 100 \quad (\text{C.1.1})$$

where $I_{\gamma_1-2,coinc}$ is the number of γ_1 transitions observed in coincidence with γ_2 transitions, $I_{\gamma_2,singles}$ the number of γ_2 transitions in singles, thus the number of triggers and $\alpha_{CE}(\gamma_1)$ the conversion coefficient for γ_1 . The efficiency for γ_1 , ϵ_{γ_1} , is expressed in %.

C.2 Determination of efficiency using γ - γ coincidences for ^{184}Tl data set

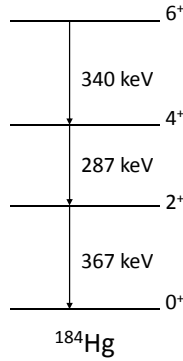
Using equation C.1.1, the detection efficiencies have been determined for a number of γ transitions in ^{184}Hg , fed in the β decay of ^{184}Tl . An overview is given in Tables C.1 and C.2 for Ge1 and Ge2 respectively. The efficiencies have been determined for the surface ionization (SI) data set, where the thallium atoms were ionized only through surface ionization and for the laser ionized (LI) data set, where the first excitation step laser frequency is tuned in order to selectively enhance the production of the (10^-) state and, through its IT, also the (7^+) state relative to the (2^-) state. The placement of the different γ transitions in the ^{184}Hg partial level scheme is depicted in Fig. C.1.

Table C.1: Detection efficiency for Ge1 from efficiency calibration and using γ - γ coincidences for ^{184}Hg .

Data set	E_{γ_1} [keV]	E_{γ_2} [keV]	$\epsilon_{calib,\gamma_1}$ [%]	$\epsilon_{coinc,\gamma_1}$ [%]
SI	367	287	7.28(16)	1.98(59)
	367	340	7.28(16)	1.94(58)
	287	340	8.51(25)	2.03(61)
LI	367	287	7.28(16)	2.22(67)
	367	340	7.28(16)	2.00(60)
	287	340	8.51(25)	2.42(73)

Table C.2: Detection efficiency for Ge2 from efficiency calibration and using γ - γ coincidences for ^{184}Hg .

Data set	E_{γ_1} [keV]	E_{γ_2} [keV]	$\epsilon_{calib,\gamma_1}$ [%]	$\epsilon_{coinc,\gamma_1}$ [%]
SI	367	287	0.99(2)	0.76(23)
	367	340	0.99(2)	0.75(25)
	287	340	1.07(3)	0.81(24)
LI	367	287	0.99(2)	0.76(23)
	367	340	0.99(2)	0.71(21)
	287	340	1.07(3)	0.76(23)

Figure C.1: Partial level scheme of ^{184}Hg , including the 367, 287 and 340 keV transitions.

From the results in Tables C.1 and C.2, it is seen that the effect of the high multiplicity is roughly the same for all γ transitions using the same detector, i.e. the efficiency curve is shifted relative to the efficiency curve obtained using the standard calibrated sources. Based on these results, it can be concluded that, within the error, it is not necessary to take into account the difference in multiplicity between the individual γ transitions and that the relative efficiencies based on the standard calibrated sources can be used in the analysis of the β decay of ^{184}Tl .

It should be noted that the reduction in absolute efficiency is comparable for both the SI and LI data sets.

C.3 Determination of efficiency using γ - γ coincidences for ^{182}Tl data set

Using equation C.1.1, the detection efficiencies have been determined for a number of γ transitions in ^{182}Hg , fed in the β decay of ^{182}Tl . An overview is given in Tables C.3 and C.4 for Ge1 and Ge2 respectively. The difference in effective efficiency for $^{182,184}\text{Tl}$ is due to the higher multiplicity in the β decay of ^{182}Tl compared to ^{184}Tl . The placement of the different γ transitions in the ^{182}Hg partial level scheme is depicted in Fig. C.2.

Table C.3: Detection efficiency for Ge1 from efficiency calibration and using γ - γ coincidences for ^{182}Hg .

E_{γ_1} [keV]	E_{γ_2} [keV]	$\epsilon_{calib,\gamma_1}$ [%]	$\epsilon_{coinc,\gamma_1}$ [%]
351	261	7.50(18)	0.77(23)
351	332	7.50(18)	0.86(26)
261	332	8.98(28)	0.97(29)

Table C.4: Detection efficiency for Ge2 from efficiency calibration and using γ - γ coincidences for ^{182}Hg .

E_{γ_1} [keV]	E_{γ_2} [keV]	$\epsilon_{calib,\gamma_1}$ [%]	$\epsilon_{coinc,\gamma_1}$ [%]
351	261	1.01(2)	0.44(13)
351	332	1.01(2)	0.45(14)
261	332	1.10(3)	0.50(15)

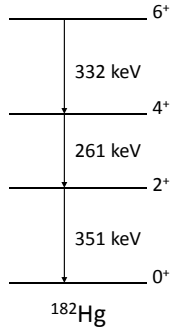


Figure C.2: Partial level scheme of ^{182}Hg , including the 351, 261 and 332 keV transitions.

C.4 Discussion

In Fig. C.3 and C.4 the detection efficiency for Ge1 and Ge2, respectively, from the standard calibrated sources (black squares) and using γ - γ coincidences for ^{182}Tl (red circles) and ^{184}Tl (green triangles, hollow for SI and full for LI) are depicted. As expected is the reduction in efficiency smaller relative to the calibration values smaller for Ge2 than Ge1, since Ge2 is placed at a larger distance from the carbon foil, this minimizing the simultaneous detection of two or more γ rays from the same decaying nucleus. It is clear that the reduction in efficiency relative to the calibration values, for both Ge1 and Ge2, is larger for ^{182}Tl , which indicates a higher multiplicity in the β decay of ^{182}Tl compared to ^{184}Tl . A detailed description about summing effects in close-detector configurations and the effect on the efficiency is given in the PhD thesis of J. Van Roosbroeck [135].

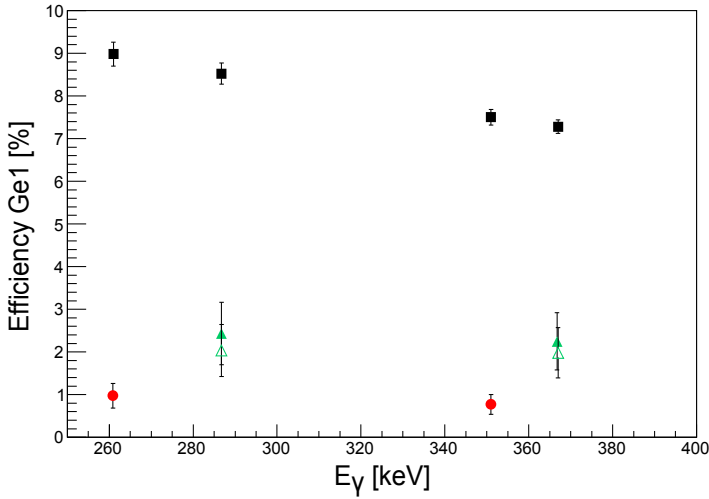


Figure C.3: Detection efficiency for Ge1 from the standard calibrated sources (black squares) and using γ - γ coincidences for ^{182}Tl (red circles) and ^{184}Tl (green triangles, hollow for SI and full for LI).

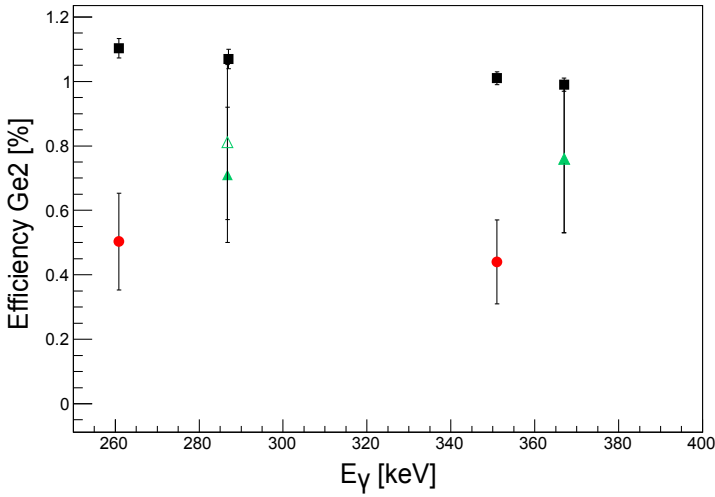


Figure C.4: Detection efficiency for Ge2 from the standard calibrated sources (black squares) and using γ - γ coincidences for ^{182}Tl (red circles) and ^{184}Tl (green triangles, hollow for SI and full for LI).

D | Intensity of coincident γ rays in $^{182,184}\text{Tl}$ α decay

This appendix gives the intensities of the γ rays observed in coincidence with the different α decays from $^{182,184}\text{Tl}$ and is an addition to the information given in section 4.3 of chapter 4.

D.1 ^{184}Tl

D.2 ^{182}Tl

Table D.1: Overview of ^{184}Tl α decay, including the different isomers and corresponding α -decay branching ratio b_α , energy E_α , intensity I_α from purified singles spectra (Si1-2), relative intensity I_α from purified singles spectra (Si1-2), reduced α -decay width δ_α^2 and γ rays and their intensity observed in coincidence (Si1-2 with Ge1). The sum $Q_\alpha + E_\gamma$ is also given for a number of transitions. The uncertainties are indicated between brackets and include both statistical and systematic contributions.

J^π , b_α (%)	E_α (keV)	I_α (counts)	$I_{rel,\alpha}$ (%)	δ_α^2 (keV)	E_γ (keV)	I_γ (counts)	$Q_\alpha + E_\gamma$ (keV)
(10^-) , 0.089(19)	6132(19) ^a	3181(60)	100	16(4)	205.9(2)	20(4)	6474(19)
					162.6(1)	38(6)	
					107.9(2)	26(5)	
					101.3(6)	23(5)	
					$\sim 78^e$	32(10)	
(7^+) , 0.047(6)	5659(10) ^a	1678(49)	100	4.9(12)	Au K_β	31(4)	
					Au K_α	82(9)	
					363.3(6)	6(2)	6148(10)
					261.8(3)	15(4)	
					257.7(3)	16(4)	
					175.7(3)	17(4)	
					$\sim 78^e$	23(8)	
					Au K_β	23(3)	
(2^-) , 1.22(30)	$\left\{ \begin{array}{l} 6161^b \\ 5988(12) \\ +5964(12)^c \\ 5964(13)^a \\ 5988(12)^a \end{array} \right.$	33279(360) ^f	93(1)	0.57(6)			6298
			100	2.4(4)			
		5810(12) ^a	6.0(3)	0.9(1)			
					224.3(3)	7(3)	6321(13)
					201.3(3)	18(4)	6322(12)
					184.2(1)	41(6)	6305(12)
					178.5(1)	51(7)	
					126.3(1)	186(14)	
					Au K_β	193(14)	
					Au K_α	423(21)	
					365.1(2)	47(7)	6304(12)
					315.1(2)	8(3)	
					272.8(3)	30(5)	
					198.4(9)	15(4)	
					Au K_β	27(5)	
					Au K_α	58(8)	
	5748(12) ^a	<128 ^f	<0.4	<0.09	426.0(5)	4(2)	6302(12)

^a E_α from α - γ coincidences

^b E_α used to calibrate the silicon detectors

^c Dominant contribution from 5988(12) α line

^d I_α for range indicated by vertical dashed lines in Fig. 4.16

^e Determined by subtracting the Au K_β intensity, calculated from the Au K_α -line intensity

^f Intensity using all data, not only the data where the wheel was turning. This was done in order to be able to compare it with the coincident γ -ray intensity, also determined using all data

Table D.2: Overview of ¹⁸²Tl low-spin α decay, including the α -decay branching ratio b_α , energy E_α , intensity I_α (Si2), relative intensity I_α (Si2), reduced α -decay width δ_α^2 and γ rays and their intensity observed in coincidence (Si2 with Ge1). The sum $Q_\alpha + E_\gamma$ is also given for a number of transitions. The uncertainties are indicated between brackets and include both statistical and systematic contributions.

b_α (%)	E_α (keV)	I_α (counts)	$I_{rel,\alpha}$ (%)	δ_α^2 (keV)	E_γ (keV)	I_γ (counts)	$Q_\alpha + E_\gamma$ (keV)
>0.49	6406 ^a	9367(97)	21(3)	>0.017			6550
	6360(6)	7301(85)	16(3)	>0.019			6503(6)
	6165(6) ^b	28080(168)	62(10)	>0.45	247.2(5)	13(4)	6551(6)
					232.2(1)	299(18)	
					197.5(2)	66(9)	6501(6)
					182.3(2)	175(15)	
					169.2(1)	230(16)	
					132.9(4)	22(6)	
					118.7(3)	44(15)	
					112.9(2)	53(12)	
					Au K β	102(17)	
					Au K α	241(20)	
	6046(5) ^b	45500(213)	100	>2.3	361.5(1)	92(10)	6543(6)
					312.6(1)	92(10)	
					296.7(3)	34(6)	
					247.8(7)	30(9)	
					231.8(2)	154(14)	
					197.5(8)	28(8)	
					182.7(3)	120(2)	
					169.3(2)	116(16)	
					153.4(2)	126(16)	
					131.7(4)	70(17)	
					112.9(1)	346(23)	
					102.0(5)	60(16)	
					Au K β	253(22)	
					Au K α	529(27)	
	5962(5) ^b	20520(143)	45(7)	>2.4	446.1(14)	10(4)	6542(5)
					265.8(2)	26(5)	
					232.2(3)	31(7)	
					206.7(1)	39(7)	
					169.2(3)	39(11)	
					112.9(1)	201(16)	
					Au K β	125(14)	
					Au K α	232(18)	

^a E_α used to calibrate the silicon detectors

^b E_α from α - γ coincidences

E | Overview of α -decay energy spectra collected in IS511.

In this appendix an overview is given of the singles α -decay energy spectra collected in Si2 in IS511. The most intense α lines are indicated with the isotope to which they belong. Data are from [8] and this work.

E.1 Mass 179

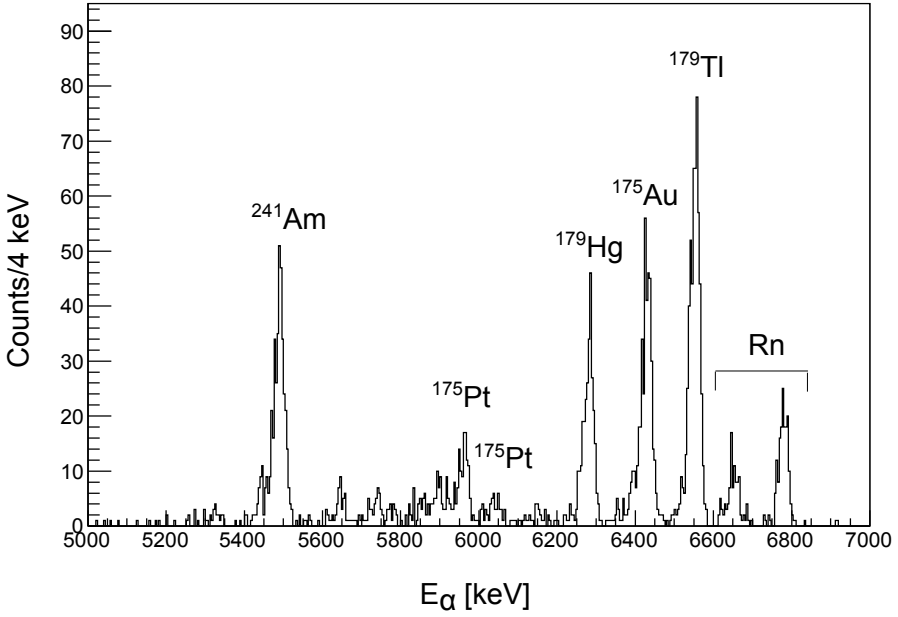


Figure E.1: Relevant part of the singles α -decay energy spectrum collected at the implantation position (Si2) for mass 179. The α peaks are denoted with the isotope to which they belong.

E.2 Mass 180

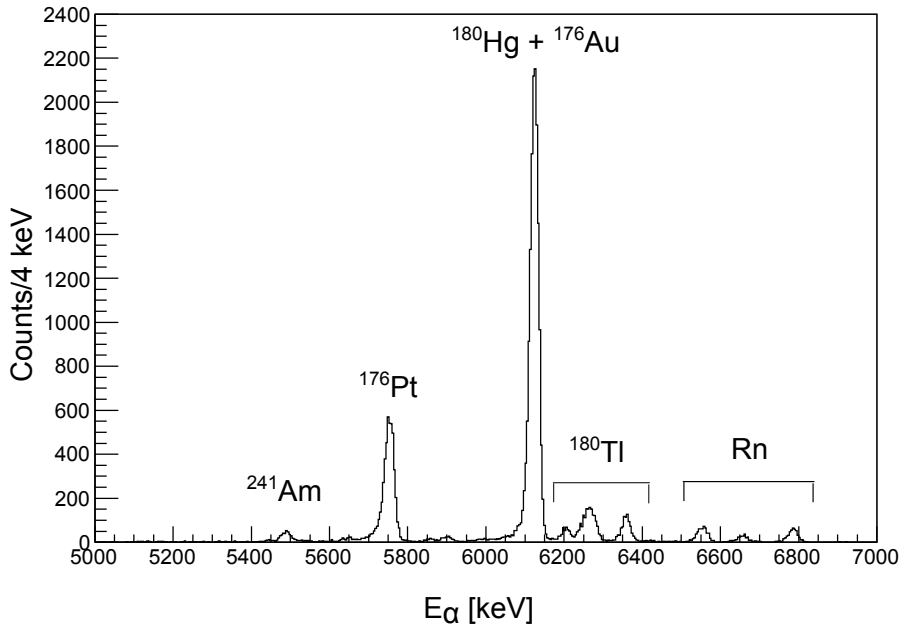


Figure E.2: Relevant part of the singles α -decay energy spectrum collected at the implantation position (Si2) for mass 180. The α peaks are denoted with the isotope to which they belong.

E.3 Mass 181

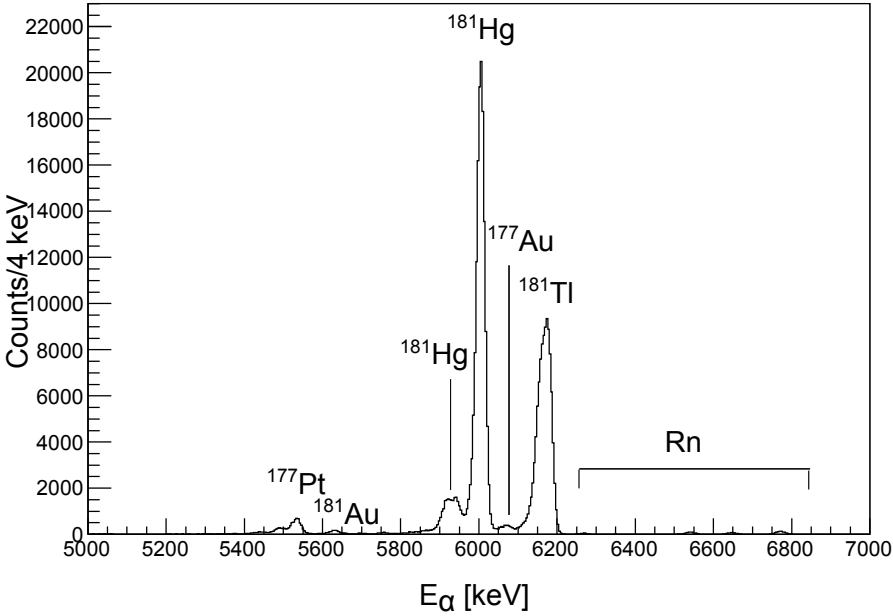


Figure E.3: Relevant part of the singles α -decay energy spectrum collected at the implantation position (Si2) for mass 181. The α peaks are denoted with the isotope to which they belong.

E.4 Mass 182

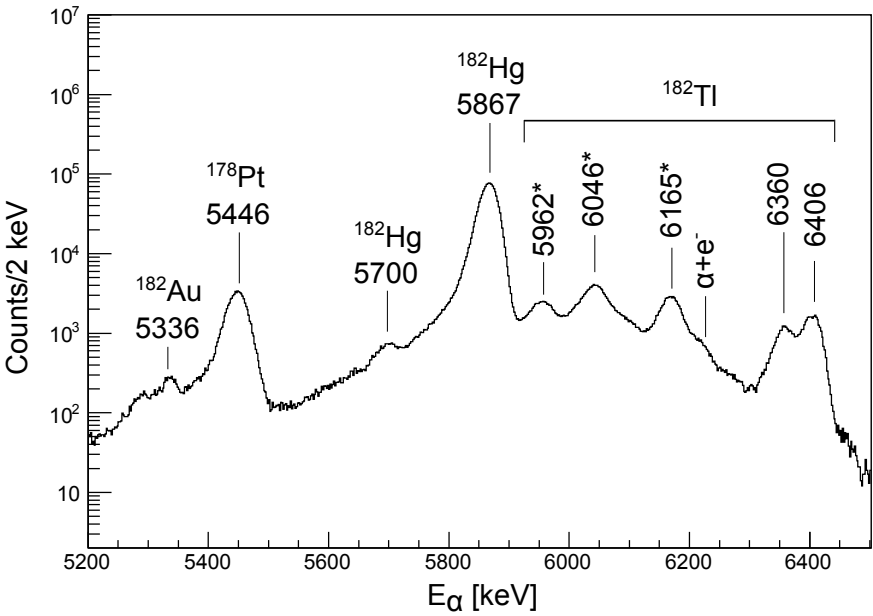


Figure E.4: Relevant part of the singles α -decay energy spectrum at the implantation position measured at mass 182 (Si2). The α peaks are denoted with the isotope to which they belong. (*: energy determined from α - γ coincidences)

E.5 Mass 183

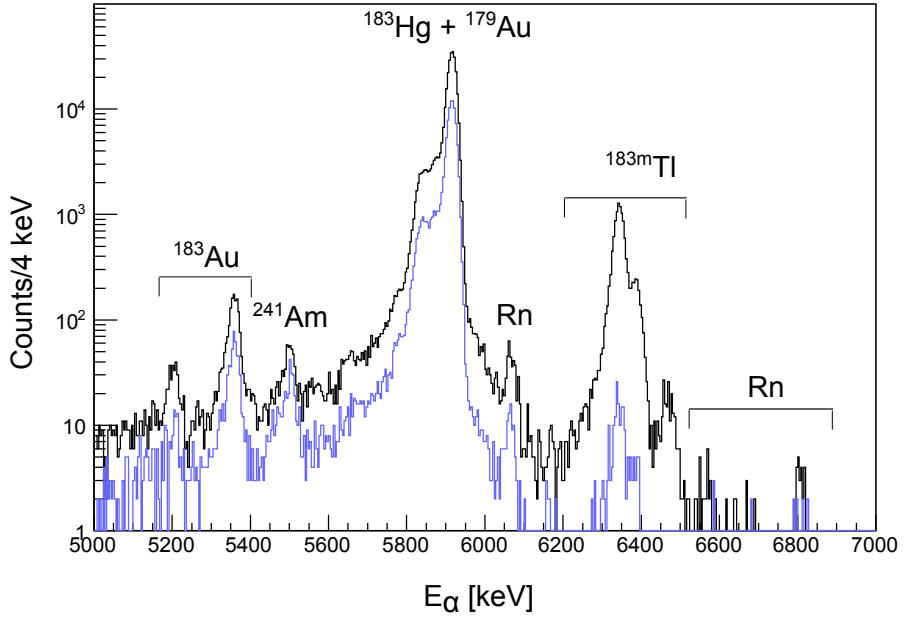


Figure E.5: Relevant part of the singles α -decay energy spectrum collected at the implantation position (Si2) for mass 183. The α peaks are denoted with the isotope to which they belong. The black spectrum corresponds to measurements in i.s. mode, the blue spectrum to measurements in g.s. mode.

E.6 Mass 184

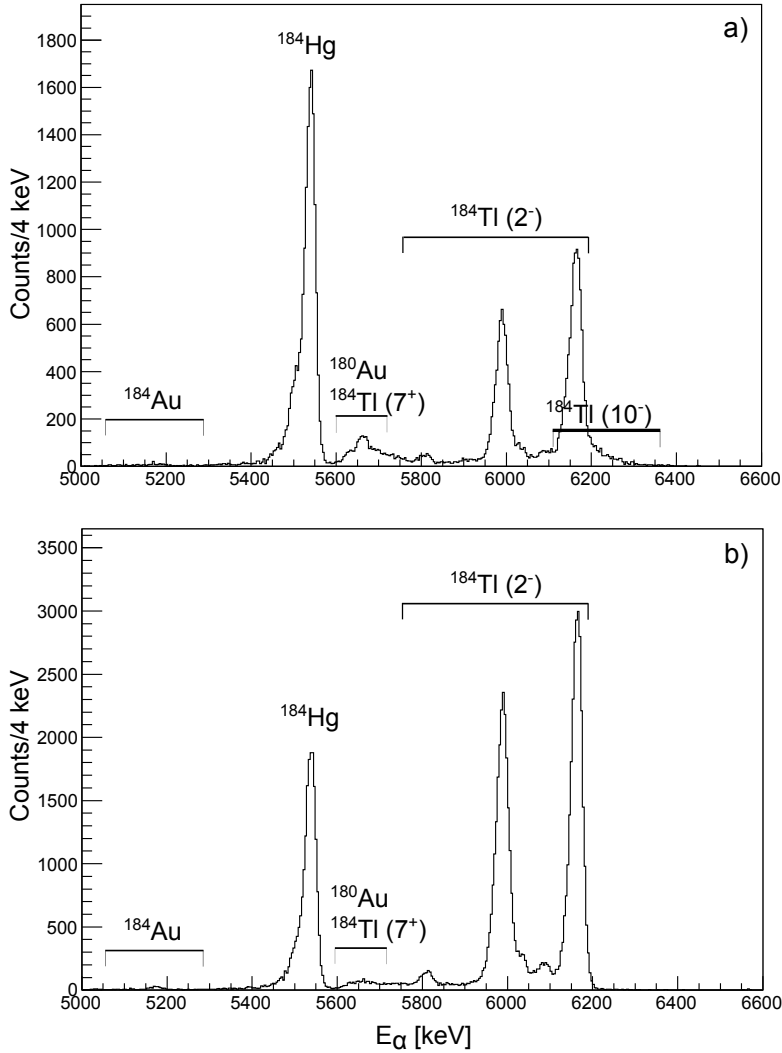


Figure E.6: Relevant part of the singles α -decay energy spectrum collected at the implantation position (sum of Si1 and Si2). Spectra a) and b) correspond to the LI and SI data sets respectively (see chapter 4). The α peaks are denoted with the isotope (and possible spin) to which they belong.

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