

In-Source laser ionization spectroscopy of $^{181,182}\text{Hg}$

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Scientific summary

The mean square charge radii of the mercury isotopes exhibit a large odd-even staggering for the neutron-deficient mercury isotopes, which is suggested to be a manifestation of shape coexistence. Shape coexistence is the phenomenon in which two (or more) distinct types of deformation occur at low energy in one and the same atomic nucleus. Understanding the occurrence of shape coexistence in atomic nuclei is a challenge faced by theories of nuclear structure. The investigation of the neutron-deficient mercury isotopes ($Z = 80$) plays an important role in the understanding of shape coexistence in this region of the nuclear chart. The theoretical description of shape coexistence is discussed in chapter 1.

In a recent laser-spectroscopy experiment performed at the ISOLDE facility in CERN, the hyperfine spectra were measured for 15 different mercury isotopes ranging from ^{177}Hg to ^{208}Hg . One of the main goals of this experiment was to extend the currently existing knowledge of the mean square charge radius behavior down towards the more neutron-deficient isotopes and verify if the mercury isotopes become spherical again or whether the more neutron-deficient nuclei become even more deformed.

In this work the difference in mean square charge radii between $^{181,182}\text{Hg}$ and ^{198}Hg are determined and compared with the literature values to verify consistency. The mercury isotopes were produced via a spallation reaction induced by a collision of accelerated protons with a molten lead target. The mercury isotopes were ionized using a three-step laser ionization process after which they were mass separated and sent to a detection system. ^{198}Hg was detected using a Faraday cup or the Multi-reflection Time-Of-Flight set-up with a multichannel-plate ion detector, while the $^{181,182}\text{Hg}$ isotopes were sent to the Windmill system. In this set-up the isotopes are implanted in carbon foils and the alpha decay was detected using silicon detectors. The aim of the experiment was to construct the hyperfine spectra of which the isotope shift and change in mean square charge radius can be determined, as explained in chapter 2. The experimental set-up is explained in chapter 3. The final results are discussed in chapter 5.

A key element in the laser spectroscopy measurements is the precise determination of the laser wavelength. However, during the past year a number of problems were identified in two wavelength meter models that are widely used at several laser laboratories. As one of the wavelength meters was of the same type as the one used for the mercury laser spectroscopy measurements, a series of systematic measurements to characterize the performance of the wavelength meters were performed in the In-Gas Laser Ionization and Spectroscopy (IGLIS) laser laboratory. The discrepancy has been resolved and it can be concluded that the charge radii measured with the in-source laser ionization spectroscopy technique are reliable.

Summary in layman's terms

All the material surrounding us is built of atoms. An atom consists of an electron cloud moving around a nucleus. The nucleus consists of protons and neutrons, which are the so-called nucleons. The chemical element is determined by the number of protons. Nuclei with the same amount of protons, but a different amount of neutrons are called isotopes.

In previous experimental work the shape of the ground state, which is the lowest energy state, of the mercury isotopes was investigated for $^{181-206}\text{Hg}$. It was observed that the ground state has a spherical shape for all isotopes except for $^{181,183,185}\text{Hg}$. In those isotopes the ground state has a “baseball-like” shape. This behaviour was interpreted as a manifestation of shape coexistence. Shape coexistence is the phenomenon in which a nucleus can have a different shape if one adds a bit of energy. In April 2015 measurements of the shape of more neutron-deficient mercury isotopes were performed in order to investigate if the nucleus would become spherical again or even more deformed. These measurements are essential to understand the occurrence of shape coexistence in atomic nuclei, which is nowadays a challenge faced by theoretical nuclear physicists.

The mercury isotopes were produced by collisions of high-energetic protons with a molten lead target. After the production, the mercury isotopes were exposed to laser light of three different frequencies. The first laser brings the atoms from the ground state into a first excitation level, the second laser excites the atoms further to a second excitation level and the third laser ionizes the atoms once they are in the second excitation state. The frequency of the second and third laser were kept constant during the experiment, while the frequency of the first laser was scanned. Only when the energy of the first laser matches the difference in energy between the ground state and first excitation level, ions are produced and sent to the Windmill set-up in which their radioactive decay is detected. The number of ions produced at a particular laser frequency could be determined by counting the number of alpha particles from the radioactive decay of mercury at this frequency. This results in a graph in which the number of ions are plotted in function of the frequency of the first laser. From this spectrum, the frequency of the first transition can be derived. The energy of this transition differs from one isotope to another, this is the so-called isotope shift and can be correlated to the change in ground state shape between the isotopes.

In this work the isotope shift between $^{181,182}\text{Hg}$ and ^{198}Hg is determined and compared with the values observed in earlier experiments to verify consistency.

A key element in the laser spectroscopy measurements is the precise determination of the laser wavelength. However, during the past year a number of problems were identified in two wave-

length meter models that are widely used at several laser laboratories. As one of the wavelength meters was of the same type as the one used for the mercury laser spectroscopy measurements, a series of systematic measurements to characterize the performance of the wavelength meters were performed in the In-Gas Laser Ionization and Spectroscopy (IGLIS) laser laboratory.

List of abbreviations

β -RADOP	β -radiation detected optical pumping method
μ BG	Micro beam gate
ABU	Atomic Beam Unit
Au	Gold
CERN	European Center for Nuclear Research
COG	Center of gravity
CW	Continuous Wave
EC	Electron capture
FC	Faraday cup
FSR	Free Spectral Range
FWHM	Full width at half maximum
GPS	General Purpose Separator
HELIOS	Heavy Element Laser Ionization Spectroscopy
HFS	Hyperfine spectra
HPGe	High-Purity Germanium
HRS	High Resolution Separator
Hg	Mercury
IBM	Interacting boson model approximation
IBM-CM	Interacting boson model with configuration mixing
IC	Internal conversion
IGLIS	In-Gas Laser Ionization and Spectroscopy
IKS	Instituut voor Kern- en Stralingsfysica
IS	Isotope shift
ISOL	Isotope separation on line
Ir	Iridium

LEGe	Low Energy Germanium
MCP	Multi-Channel Plate
MR TOF	Multi-reflection Time-Of-Flight
Mle	Maximum likelihood estimation
Os	Osmium
PIPS	Passivated Implanted Planar Silicon
PSB	Proton Synchrotron Booster
Pb	Lead
Pt	Platinum
RFQ	Radiofrequency quadrupole
RILIS	Resonance Ionization Laser Ion Source
Rms	Root mean square
SATLAS	Statistical Analysis Toolbox for Laser Spectroscopy
SC	Super cycle
SFPI	Scanning Fabry-Pérot Interferometer
SMS	Specific mass shift
TBME	Two-body matrix elements
TOF	Time of Flight
WM	Windmill

List of symbols

A	Mass number
Z	Number of protons
N	Number of neutrons
H	Hamiltonian
ψ	Wavefunction
$\langle r^2 \rangle$	Mean square charge radius
$\langle \beta^2 \rangle^{1/2}$	Rms quadrupole deformation parameter
β	Quadrupole deformation parameter/beta
I	Nuclear spin
L	Electron orbital angular momentum quantum number
S	Electron spin quantum number
J	Total electron angular quantum number
F	Total atomic angular quantum number
$\delta \langle r^2 \rangle^{AA'}$	Change in mean square charge radius between isotope A and A'
μ	Magnetic dipole moment
Q_s	Spectroscopic quadrupole moment
$\delta \langle v^2 \rangle^{AA'}$	Isotope shift between isotope A and A'
χ^2	Chi-Square
χ_{red}^2	Reduced Chi-Square

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Chapter 1

Motivation

This first chapter starts with a general introduction to nuclear structure and decay properties. Also different theoretical nuclear models relevant for the nuclei studied in this work are briefly explained. The chapter concludes by giving the motivation for studying mercury isotopes as well as a short discussion of the theoretical considerations.

1.1 Nuclear structure

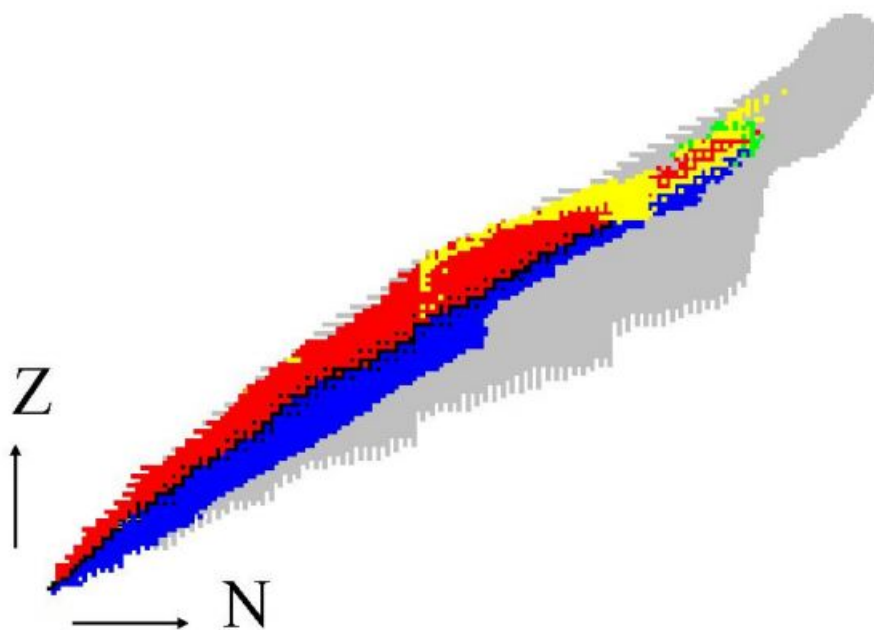


Figure 1.1: The nuclear chart with the number of neutrons N on the horizontal axis and the number of protons Z on the vertical axis [1]. The nuclei represented in grey have not yet been observed. The colors denote the different decay processes. (Red: β^+ decay/electron capture. Blue: β^- decay. Yellow : α decay. Green: spontaneous fission.)

Atoms are the building blocks of everything around us, including ourselves. An atom consists of a nucleus surrounded by an electron cloud. The nucleus is composed of positively charged protons and uncharged neutrons, which are called nucleons. The chemical

element is determined by the number of protons, represented by the atomic number Z . Two nuclei with the same number of protons but with a different amount of neutrons N are called isotopes of that particular element. The total number of protons and neutrons are expressed by the mass number A ($= Z + N$). The notation ${}^A\text{X}$ is used to indicate a particular nucleus with A nucleons and Z protons, with X representing the name of the element. The nuclear chart, shown in figure 1.1, displays all the theoretical predicted nuclei (around 6000 nuclei) as a function of their proton number Z and neutron number N .

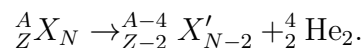
Today, more than 3600 nuclei have been experimentally observed of which only 284 are stable. A stable nucleus has a time scale for possible radioactive decay of the order of the time scale of the universe or even longer [2]. The remaining nuclei are in an unstable state and will decay to other, more stable nuclei. The stability of a nucleus is determined by the balance between all interactions at play within the nucleus, which includes the strong, electromagnetic and weak interaction, while gravitation is neglected at this scale. The strong nuclear force acts on the femtometer scale and can hold the nucleons together provided that the amount of neutrons is sufficient. The ratio of the neutron number to the proton number N/Z determines if the strong force can overcome the electromagnetic repulsion between the protons. Therefore this ratio plays a major role in the stability of the nucleus. Besides the strong and electromagnetic force, the weak interaction is another important force since it drives the beta decay of nuclei. Stable nuclei lie on the so-called “beta stability line”. As can be seen on the nuclear chart, this line follows the $N = Z$ trend for light nuclei and deflects more towards the neutron-rich side for heavy nuclei. Nuclei with an extreme N/Z ratio (compared to the N/Z ratio of stable nuclei) are referred to as “exotic nuclei”.

1.2 Nuclear decay processes

The most important nuclear decay processes are α decay, β decay and γ decay. A process competing with γ decay is internal conversion (IC). The most important decay process of a certain nucleus is indicated in figure 1.1.

1.2.1 α decay

A nucleus undergoing alpha decay emits an α particle, which is a helium core (${}^4_2\text{He}$) consisting of two protons and two neutrons. This process is represented as



The emitted α particles have well-defined energies since there are only two decay products. Alpha emission can be considered a quantum tunneling process in which the α particle has to tunnel through the Coulomb barrier. A nucleus is an α emitter if a net energy is released during the process and if the probability to tunnel through the Coulomb barrier is not too small. (Otherwise α emission may occur so rarely that it may not be detected [3].)

1.2.2 β decay

β decay is governed by the weak interaction. The three known β -decay processes are β^- decay, β^+ decay and electron capture (EC). In all of these processes the number of nucleons is conserved while a proton is converted into a neutron or vice versa. In β^- decay a neutron is converted to a proton together with an electron e^- and anti-neutrino $\bar{\nu}$, written as

$${}^A_Z X_N \rightarrow {}^A_{Z+1} X'_{N-1} + e^- + \bar{\nu}.$$

β^+ decay on the other hand converts a proton into a neutron, thereby emitting a positron e^+ and neutrino ν :

$${}^A_Z X_N \rightarrow {}^A_{Z-1} X'_{N+1} + e^+ + \nu.$$

The released β particles have a continuous energy distribution due to the fact that $\beta^\mp$ decay is a three body process. The emitted positron can annihilate with a surrounding electron once it is slowed down. This results in the creation of two γ rays emitted back to back with an energy of 511 keV. A competing process of β^+ decay is EC. In EC an atomic electron, mostly from the K- or L-shell, is swallowed by the nucleus allowing a proton to be converted to a neutron and a neutrino ν :

$${}^A_Z X_N + e^-_{\text{orbital}} \rightarrow {}^A_{Z-1} X'_{N+1} + \nu.$$

After this process the atom is in an excited state with a vacancy in an electron orbital. Therefore the atomic electrons will rearrange themselves and characteristic X-rays or Auger electrons are emitted.

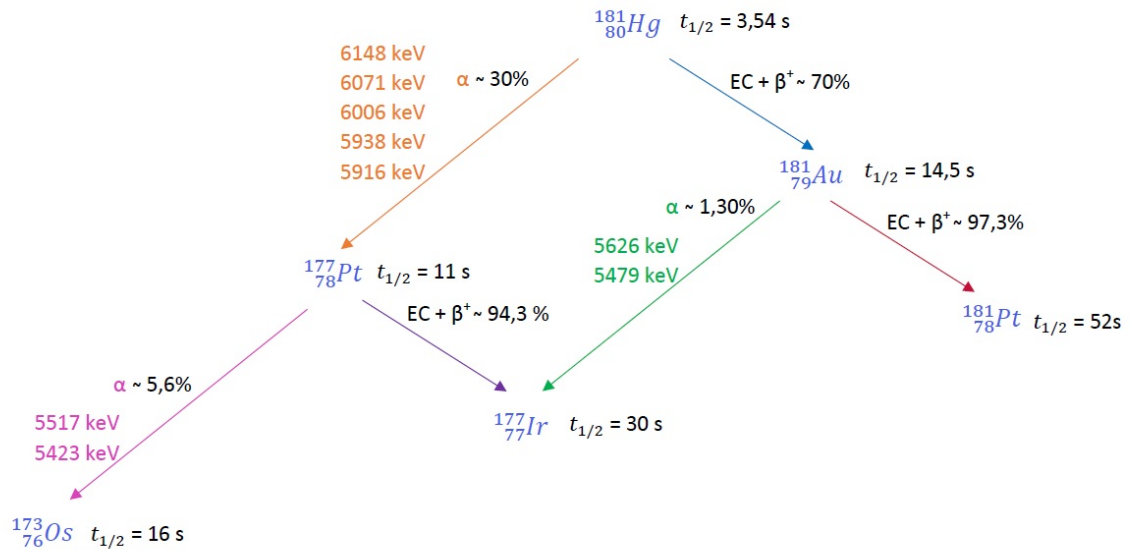
1.2.3 γ decay

The α decay and β decay processes can leave the daughter nucleus in an excited state. This state can subsequently decay through gamma emission which is mediated by the electromagnetic force. In this process a high energy photon is emitted which matches the change in energy and orbital angular momentum between the two involved states of the nucleus. The excess energy can also be given to an atomic electron via internal conversion. The conversion electrons are emitted with a specific energy depending on the energy difference between the two involved states of the nucleus and the atomic binding energy of the electron. After internal conversion a vacancy is left in an electron orbital. The vacancy is filled by other electrons thereby emitting characteristic X-rays and Auger electrons.

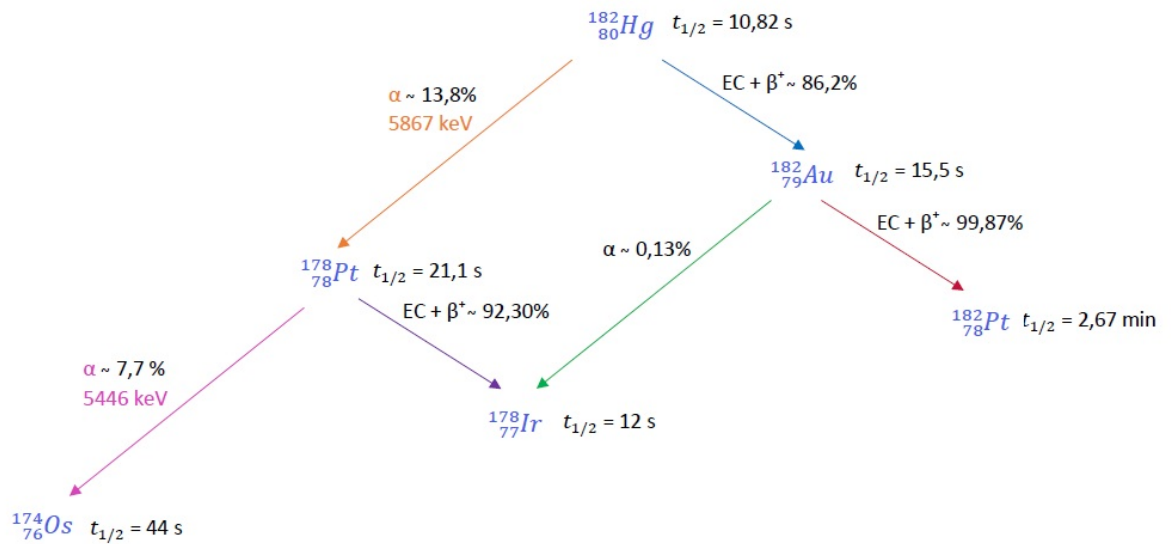
1.2.4 Decay scheme of $^{181,182}\text{Hg}$

In this work laser spectroscopy measurements to obtain differences in charge radii are reported. The hyperfine structure spectra are made and analyzed for the isotopes $^{181,182}\text{Hg}$. The decay scheme of these isotopes [4] is shown in figures 1.2a and 1.2b.

Every nucleus in the decay scheme undergoes either α -decay or electron capture (EC) and β^+ -decay. Each decay step is represented by a specific color e.g. the energy of an alpha particle from the decay of ^{181}Hg will be shown in orange in this work and the energy of a X-/gamma ray from the β -decay of ^{181}Hg is displayed in blue. The energies of the known alpha lines are also indicated in the corresponding color.



(a)



(b)

Figure 1.2: Decay schemes of (a) ^{181}Hg and (b) ^{182}Hg . In this work the represented color code will be used to denote that an alpha or X-ray /gamma line belonging to this specific decay. The alpha decay energies are shown together with the branching ratios of the different decay branches.

1.3 Nuclear models

The Schrödinger equation $H\psi = E\psi$, with ψ the wave function describing a medium of A interaction nucleons, E an energy eigenvalue and H the Hamiltonian, has to be solved if one wants to understand the properties of different nuclei. The non-relativistic Hamiltonian H is given by:

$$H = \sum_{i=1}^A T_i + \frac{1}{2} \sum_{i,j=1}^A V_{i,j} \quad (1.1)$$

with T_i the kinetic energy of nucleon i and $V_{i,j}$ the two-body interaction potential energy between nucleon i and j . Higher order terms (e.g. three-body forces) originating from the neglect of the internal degrees of freedom of the nucleons (nucleons are build from quarks) are omitted in formula 1.1. Analytically solving this Schrödinger equation is only possible for the lightest nuclei and the exact form of the nucleon-nucleon interaction force is still poorly known. Therefore some simplifications/approximations are made to describe a nucleus.

Theoretical nuclear models can be divided into several categories depending on how they deal with the complex many-body problem of the nucleus. A theory is either a macroscopic (e.g. Liquid-drop model and droplet model), a microscopic (e.g. Shell model, Nilsson model and mean field approaches) or a macroscopic-microscopic model (e.g. Strutinsky shell-correction method). The various nuclear models of interest in this work are briefly described in the next subsections.

1.3.1 Macroscopic models

Liquid-drop model

Electron scattering experiments gave the insight that the nuclear density distribution is roughly constant [3]. This served as an inspiration for the liquid-drop model. In this model the protons and neutrons are homogeneously distributed over the volume of the nucleus, which is considered a sphere with radius $R = r_0 A^{1/3}$ where r_0 equals 1.2 fm. The liquid-drop model is based on similarities between such a nucleus and a drop of incompressible fluid. The semi-empirical mass formula was derived with the aid of the liquid-drop model in combination with some shell model effects in order to calculate the binding energy $B(Z, A)$ of a nucleus with A nucleons and Z protons. The binding energy $B(Z, A)$ is given by:

$$B(Z, A) = a_v A - a_s A^{2/3} - a_c Z(Z-1)A^{-1/3} - a_{\text{sym}} \frac{(A-2Z)^2}{A} + \delta \quad (1.2)$$

The first three terms are called the volume, surface and Coulomb term respectively and are derived from the liquid-drop model. The volume term scales with the number of nucleons A . This reflects the fact that the nuclear force is a short-range force and that every nucleon has the same amount of neighbors due to the constant nuclear density. However the nucleons at the surface are less bound because they have less neighbors. This is taken into account by the surface term. The Coulomb term represents the Coulomb repulsion between the protons. The last two terms in equation 1.2 are the symmetry and pairing term based on shell model considerations. The symmetry term refers to the beta stability

line, while the pairing term represents the fact that even-even nuclei are more stable due to the pairing force.

Droplet model

The droplet model is a refinement of the liquid-drop model. The nuclear density distribution deviates from the liquid-drop picture in a few important ways. First of all, the density is no longer assumed to be constant over the nuclear volume. The Coulomb repulsion pushes the protons (and the neutrons to some extent) towards the surface. This results a.o. in an increase of all the radial moments. Secondly, the edge of the nucleus has a certain diffuseness and the effective sharp radii of the neutron and proton distributions are not considered to be equal. Instead, neutron rich nuclei have a neutron skin because the neutron distribution is slightly larger than the proton distribution [5].

The liquid-drop model does not describe the observed trends of mean square charge radii along isotonic and isotopic sequences well. The droplet model gives a more satisfactory description [6]. The mean square charge radius $\langle r^2 \rangle$ of a spherical nucleus is equal to

$$\langle r^2 \rangle = \langle r^2 \rangle_u + \langle r^2 \rangle_r + \langle r^2 \rangle_d \quad (1.3)$$

with $\langle r^2 \rangle_u = \frac{3}{5}R_z^2$ the contribution from the uniform distribution, $\langle r^2 \rangle_r = \frac{12}{175}C'R_z^2$ the contribution from the redistribution of the nucleons with $C' \approx 0.0156ZA^{-1/3}$ and $\langle r^2 \rangle_d = 3b^2$ the contribution from the diffuseness with b a droplet coefficient given in table 1.1. R_z is the sharp proton radius and is given by

$$R_z = R - \frac{N}{A}t \quad (1.4)$$

with t the neutron-skin thickness expressed by

$$t = \frac{2}{3}(I - \bar{\delta})/B_s \quad (1.5)$$

and R the combined size of protons and neutrons, which is given by

$$R = r_0A^{1/3}(1 + \bar{\epsilon}) \quad (1.6)$$

in which

$$\bar{\epsilon} = (-2a_2A^{-1/3}B_s + L\bar{\delta}^2 + c_1Z^2A^{-4/3}B_c)/K \quad (1.7)$$

$$\bar{\delta} = \frac{I + \frac{3}{16}(c_1/Q)ZA^{-2/3}B_v}{1 + \frac{9}{4}(J/Q)A^{-1/3}B_s} \quad (1.8)$$

$$I = (N - Z)/A. \quad (1.9)$$

For a spherical nucleus B_s, B_v and B_c are equal to 1 [5]. The droplet model coefficients are given in table 1.1 both for Myers and Schmidt's old [5] and Berdichevsky's new [6] parametrization form. The old parametrization form is determined by fitting to nuclear masses and fission barriers, while the new parameters were fitted to the experimental nuclear core density of ^{208}Pb .

Droplet model coefficient	Myers and Schmidt's (old) parametrization form [5]	Berdichevsky's (new) parametrization form [6]
r_0 (fm)	1.18	1.15
b (fm)	0.99	1.02
c_1 (MeV)	0.7322	0.7322
a_2 (MeV)	20.69	18
J (MeV)	36.8	29.5
Q (MeV)	17	45
K (MeV)	240	240
L (MeV)	100	-5

Table 1.1: Droplet model coefficients for both the old and new parametrization.

1.3.2 Microscopic models

Microscopic models try to find a solution of the complex Schrödinger equation describing a nucleus. However solving this equation is only possible for the lightest nuclei, for all the other nuclei assumptions have to be made to simplify the calculation. A first simplification consists of rewriting the Hamiltonian in equation 1.1 as consisting of two terms: an one-body operator (H_0) which describes the nucleus as consisting of independent nucleons moving in an average potential and a term representing the residual interaction beyond the one-body approximation (H_1). This rearrangement can be written as:

$$H = \sum_{i=1}^A \left(\frac{p_i^2}{2m} + V(\vec{r}_i) \right) + H_1 = H_0 + H_1 \quad (1.10)$$

where $V(\vec{r}_i)$ is a mean-field potential generated by the nucleons themselves and H_1 is considered as a perturbation term [7]. $V(\vec{r}_i)$ can be determined empirically or in a self-consistent way. The Wood-Saxon potential is an example of an empirical mean-field potential and serves as a starting point in the shell model. The Hartree-Fock method derives $V(\vec{r}_i)$ in a self-consistent way. This approach starts from the two-body nucleon-nucleon force and one derives an expression for $V(\vec{r}_i)$ by demanding that the energy of the ground state is minimal. This minimization is performed in an iterative way.

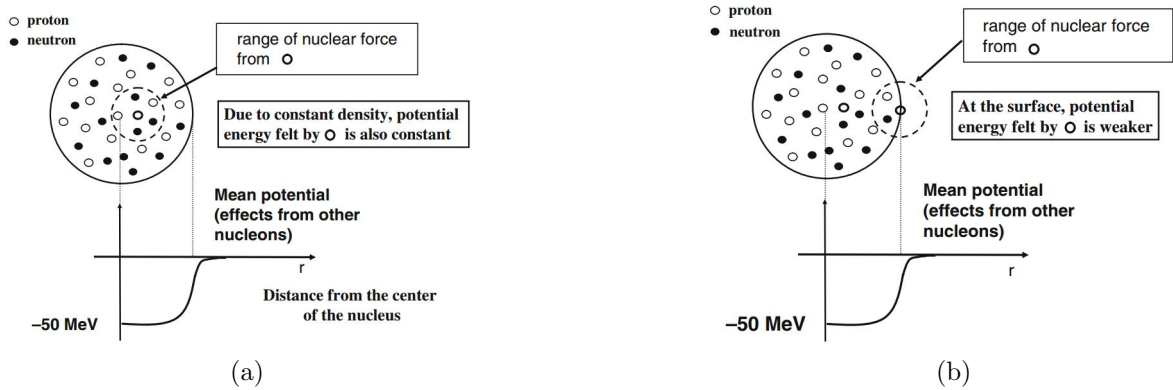


Figure 1.3: Illustration of the mean field potential field by a nucleon [8]. (a) A nucleon well inside the nucleus interacts with its neighbors within the reach of the nuclear force. Since the nuclear density is rather constant, the mean field potential should be constant as well inside the nucleus. (b) The potential energy of nucleons close or add the surface is decreasing as less nucleons interact. Therefore the mean field potential increases near the surface of the nucleus.

Shell model

Experimental evidence of increased stability for certain “magic” proton and neutron numbers (a.o. the neutron and proton separation energy) resulted in the idea that nucleons are arranged in a shell structure. Analogous to noble gas configurations in chemistry of atomic elements. Near stability the magic numbers are $N, Z = 2, 8, 20, 28, 50, 82, \dots$

The Wood-Saxon potential is used to reproduce the spherical mean field [8]. Here, it is considered that a nucleon inside of the nucleus (see figure 1.3a) interacts with the surrounding nucleons within reach of the interaction, which is about 1 fm. This is indicated by the dashed circle in figure 1.3a. Due to the constant density inside the nucleus, the mean field potential should be constant as well inside the nucleus. The potential energy of nucleons close or add the surface is decreasing as less nucleons interact (see figure 1.3b). Therefore the mean-field potential should increase gradually near the surface.

The eigenvalues of the Wood-Saxon potential can only be calculated numerically. Therefore the Wood-Saxon potential is approximated by a Harmonic oscillator potential which is simpler and can be treated analytically. If only the Harmonic oscillator potential is used to describe the mean field potential, the experimentally observed “magic” numbers near stability can not be reproduced. Mayer and Jensen could reproduce the correct magic numbers by including an attractive l^2 term and a spin-orbit coupling term ($\vec{l} \cdot \vec{s}$) with l the orbital angular momentum quantum number and s the spin quantum number. The resulting shell structure is displayed in figure 1.4. These quantized energy levels are the solutions of the Schrödinger equation of the one-body hamiltonian H_0 (equation 1.10).

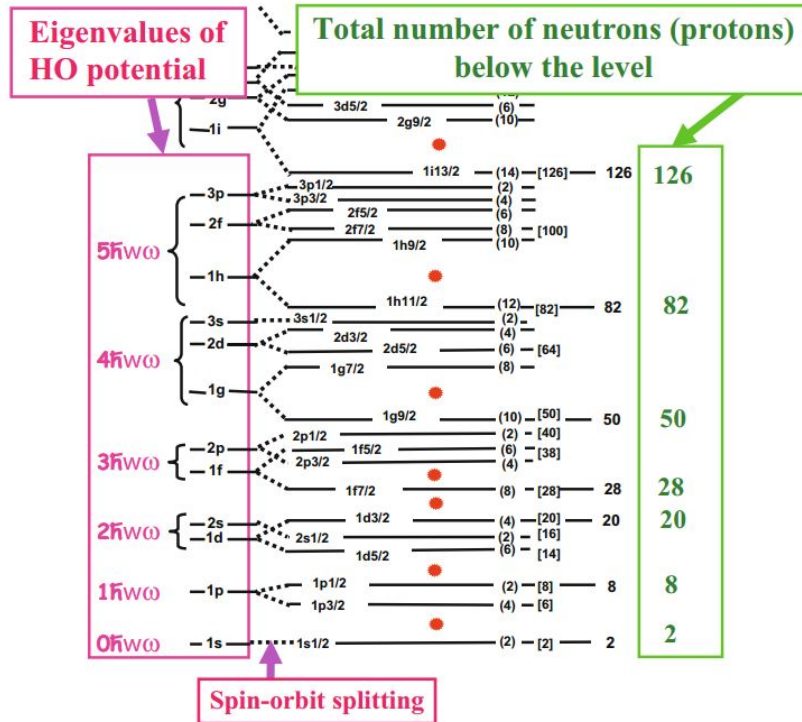


Figure 1.4: Quantized energy levels originating from a simple harmonic oscillator potential (left in pink) and from a modified harmonic oscillator potential with a l^2 and spin-orbit coupling term. Mayer-Jensen’s magic numbers are shown on the right side [8].

The magic numbers of Mayer and Jensen were derived from direct consequences of basic properties of the nuclear force and density. Therefore, the magic numbers were assumed to remain the same for all nuclei, both stable and exotic [8]. Note that at that time, only stable nuclei were experimentally investigated. Nowadays it's known that the shell structure evolves throughout the nuclear chart. In particular for nuclei far from the region of β stability, the closed shells and subshells are situated elsewhere. Consequently, the Mayer-Jensen's magic numbers are no longer valid in this region. The change of the relative distance between the single particle energies is referred to as the monopole shifts. These monopole shifts arise from proton-neutron interactions (e.g. monopole interaction and tensor interaction) of which the effects on the shell gaps become visible in the exotic region.

In the single particle picture (the Hamiltonian only consists of H_0) each nucleon is in a particular orbit. It's said that the nucleons are in a certain configuration. However, due to H_1 the orbits shown in figure 1.4 are no longer eigenstates of the total Hamiltonian H and configuration mixing occurs. This means that in general the wave function of a certain energy state of the nucleus consists of different configurations. If the energy difference between two neighboring orbits is large, including the perturbation term H_1 will result in weak mixing between those two levels. This justifies a separation of the nucleus in an inert core of nucleons sitting in a certain orbit and active valence nucleons which are allowed to “scatter” over different orbits (see figure 1.5). Due to computational limitations, the valence nucleons are only allowed to scatter in a selected set of shell model orbits. This truncation of the model space is possible due to weak mixing between levels with a large energy difference. The choice of an appropriate model space for the valence nucleons is essential in shell model calculations.

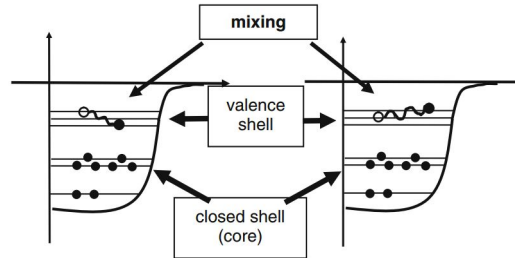


Figure 1.5: The perturbation Hamiltonian H_1 results in configuration mixing. The nucleons in the inert core are in a certain orbit, while the valence nucleons are “scattered” in different orbits [8].

In the shell model only the degrees of freedom in the valence shell are taken into account. The calculations start from a Hamiltonian which describes the energy of the valence nucleons with respect to the inert core and the residual interaction between the valence nucleons. This Hamiltonian is written in second quantization form as

$$H = \sum \epsilon_a a_a^\dagger a_a + \frac{1}{4} \sum \langle \alpha\beta | V | \gamma\delta \rangle a_\alpha^\dagger a_\beta^\dagger a_\delta a_\gamma. \quad (1.11)$$

The single-particle energies ϵ_a can be determined experimentally by proton and neutron separation energies near closed shells or from self-consistent Hartree-Fock calculations. The residual two-body interaction, expressed by means of the two-body matrix elements

$\langle \alpha \beta | V | \gamma \delta \rangle$, can be determined by fitting energy eigenvalues to experimental energies or can be derived from a fully microscopic approach using realistic forces [9]. The two-body matrix elements (TBME) take into account the nucleon-nucleon interaction in free space, to some extent the effects of three-body forces and more, the effect of excitation to orbits above the model space and the effect of neglecting the core nucleons. Therefore the TBME depend on the model space under consideration [8]. The eigenvalues of Hamiltonian H (equation 1.11) are found by diagonalizing H considering different possible configurations of the valence nucleons in the chosen model space. The off-diagonal elements resulting from the residual two-body force are responsible for configuration mixing, while the diagonal TBME are responsible for the shifts in single particle energies, the earlier mentioned monopole shifts.

Single-particle states in deformed nuclei

The energy levels of the shell model (figure 1.4) are based on the assumption of a spherical mean field potential. However this is not true for all nuclei. Some nuclei have a nonspherical equilibrium shape and are said to be deformed. Most deformed nuclei exhibit a quadrupole deformation and have either a prolate or oblate shape (figure 1.6). Both shapes are cylindrical symmetric and can be represented by an ellipsoid of revolution around the symmetry axis. In the case of a prolate shape the semimajor axis is larger than the semiminor axis of this ellipse and visa versa for an oblate shape. The surface of such a deformed nucleus is described by

$$R(\theta, \phi) = R_{av} (1 + \beta Y_{20}(\theta, \phi)). \quad (1.12)$$

The angle θ is defined with respect to the symmetry axis. This formula is independent of ϕ and therefore gives the nucleus cylindrical symmetry. The quadrupole deformation parameter β is related to the eccentricity of the ellipse as

$$\beta = \frac{4}{3} \sqrt{\frac{\pi}{5}} \frac{\Delta R}{R_{av}} \quad (1.13)$$

with $R_{av} = 1.2A^{1/3}$ fm and ΔR the difference between the semimajor and semiminor axes of the ellipse [3]. When $\beta > 0$ the nucleus is prolate deformed and when $\beta < 0$ the nucleus has an oblate shape.

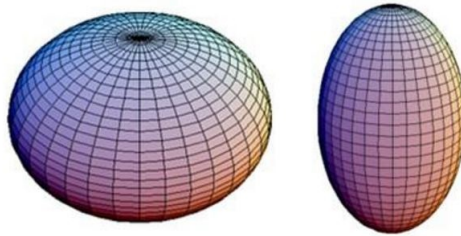


Figure 1.6: Quadrupole deformation shapes. Left: oblate shape ($\beta < 0$). Right: prolate shape ($\beta > 0$) [10].

In the case of deformed nuclei, the mean field potential is a deformed Wood-Saxon potential. S. G. Nilsson was the first who found the single-particle shell model states in a

deformed nucleus. In the so-called Nilsson model a single valence nucleon is coupled to a deformed nucleus and its single-particle Hamiltonian is given by

$$h_{\text{Nilsson}} = \frac{p^2}{2M} + \frac{1}{2}M (w_{\perp}^2(\bar{x}^2 + \bar{y}^2) + w_z^2 \bar{z}^2) + D\mathbf{l}^2 + \epsilon \mathbf{l} \cdot \mathbf{s} = h_0 + h_{\text{deformed}} \quad (1.14)$$

which is nothing else than the standard shell model Hamiltonian whereby the spherical harmonic oscillator potential is replaced by a deformed harmonic oscillator potential [11]. In the spherical case (figure 1.4), each single-particle state has a specific orbital angular momentum l , total angular momentum j and is $(2j + 1)$ -fold degenerated. In the Nilsson model l and j are no longer “good” quantum numbers due to the lack of spherical symmetry. The energy states have mixtures of different l (based on parity considerations: only odd or only even l) and j values. The corresponding wave function only contains components which have the same absolute value for the projection of j along the symmetry axis, denoted by Ω . The single-particle wave functions and energies depend on the deformation parameter β and are typically visualized in a Nilsson graph (figure 1.7) [3].

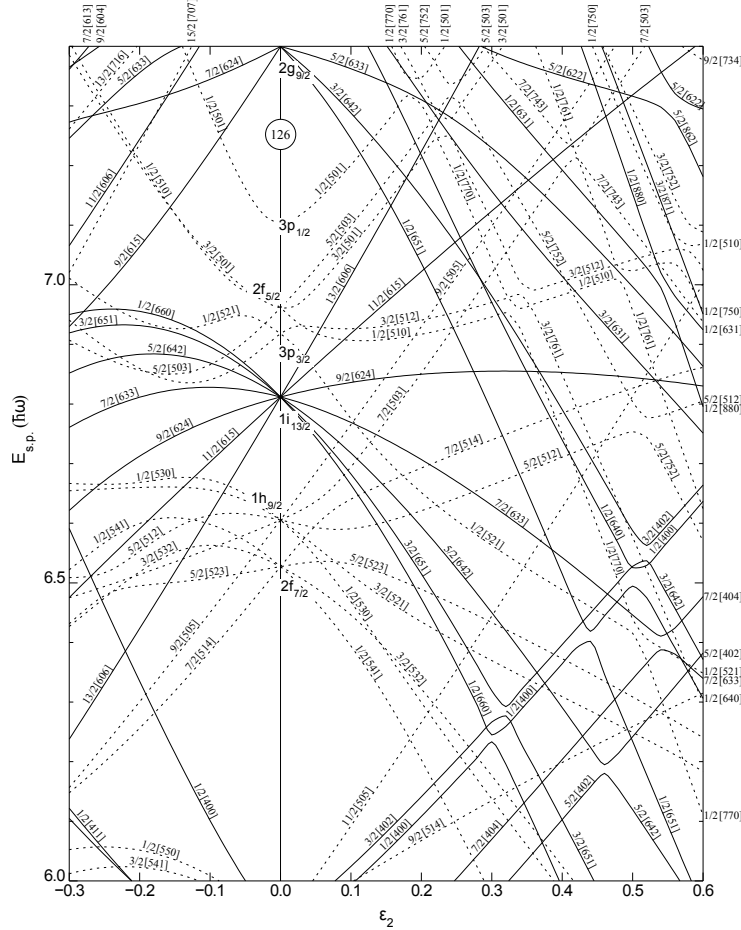


Figure 1.7: Nilsson graph. Energy levels for neutrons ($82 \leq N \leq 126$) in a prolate ($\epsilon_2 > 0$) and oblate ($\epsilon_2 < 0$) deformed nucleus for different values of the deformation ϵ_2 which is a measure of the quadrupole deformation parameter β . The levels are labeled by $\Omega [N n_z \Lambda]$ with Ω the projection of j along the symmetry axis, N the principal quantum number of the oscillator shell to which the single-particle state belongs for $\epsilon_2 = 0$, n_z the number of nodes and $\Lambda = n_z + l$ with l the orbital quantum number of the single-particle state to which the state evolves for $\epsilon_2 = 0$ [12].

In the case of small deformations ($|\beta| \approx 0$), h_{deformed} is a perturbation Hamiltonian and the eigenstates of h_{Nilsson} are calculated using perturbation theory. h_{deformed} lifts the $(2j+1)$ -fold degeneration and the energy increases or decreases linearly with deformation depending on Ω . Orbits with a considerable overlap with the deformed nucleus (small Ω for prolate shaped nuclei and large Ω for oblate deformed nuclei), interact more strongly with the nucleus and decrease in energy. Due to reflection symmetry, these states are two-fold degenerated. Once two states with the same Ω -value reach each other, two-state mixing occurs and the states “repel” each other. For ($|\beta| \gg 0$), h_{deformed} can no longer be considered a perturbation term and the total Hamiltonian h_{Nilsson} has to be diagonalized.

A deformed nucleus can rotate around an axis perpendicular to the symmetry axis. The total angular momentum $\vec{I}$ of a rotating nucleus is equal to the sum of the angular momentum of the rotating core $\vec{R}$ and the angular momentum of the valence nucleons $\vec{J}$. If one adds more energy into the rotational degrees of freedom, I increases. However the projection of I along the symmetry axis K remains the same since a nucleus can not rotate around the symmetry axis. Therefore all states within a rotational band have the same K value. The sequence of a rotational band build on the ground state of an even-even nucleus, which is always a 0^+ -state due to pairing interaction, only contains even I values (0^+ , 2^+ , 4^+ , ...). The energy of such a rotating system is given by

$$E = \frac{\hbar^2}{2\mathcal{J}} I(I+1) \quad (1.15)$$

with $\mathcal{J}$ the moment of inertia [3].

1.3.3 Macroscopic-microscopic model

Strutinsky’s shell-correction method

The Strutinsky’s shell-correction method is a microscopic-macroscopic method. It is able to give a prediction of the shape of the ground state of a nucleus. The shape is found by minimizing the deformation energy. The ground state energy is considered to be the sum of the liquid drop energy and the Strutinsky’s shell correction energy [13]. The liquid drop binding energy (equation 1.2) was calculated for a spherical shape. The surface and Coulomb term are different for a deformed nucleus and depend on the particular shape of the nucleus. Therefore the liquid drop energy is a function of the deformation parameter. The Strutinsky’s shell correction energy takes into account the change of the single-particle energy levels near the Fermi level when the nuclear shape is modified (figure 1.7). Figure 1.8 shows the results of a Strutinsky’s shell-correction method calculation in the case of odd-mass mercury isotopes. Calculations in [13] using this method expect $^{187-189}\text{Hg}$ to have a small oblate deformation while $^{181-185}\text{Hg}$ have a large prolate equilibrium deformation.

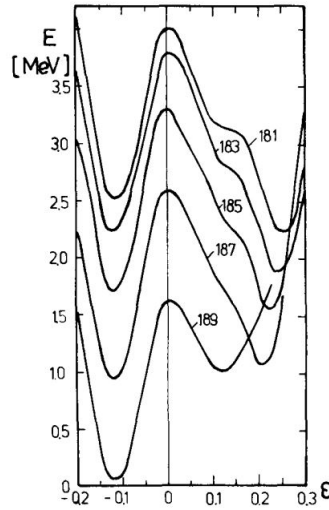


Figure 1.8: Deformation energy of the odd-mass mercury isotopes using Strutinsky's shell correction method. $^{187-189}\text{Hg}$ have a small oblate deformation, while $^{181-185}\text{Hg}$ are strongly prolate deformed [13].

1.4 Motivation to study the light mercury isotopes

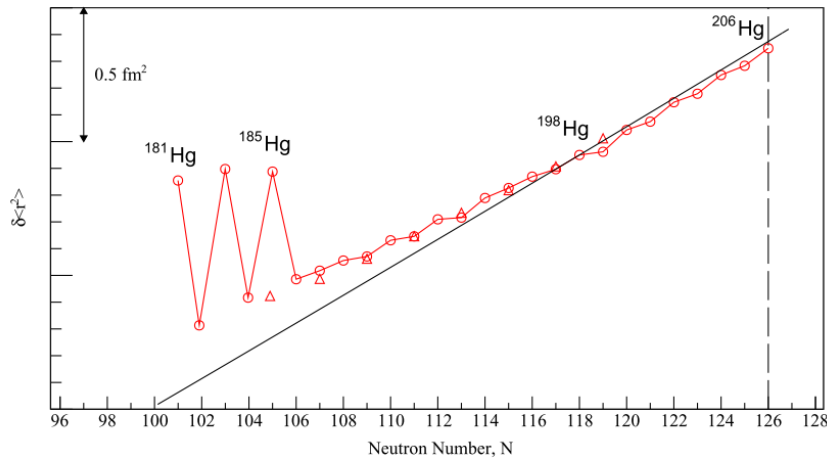


Figure 1.9: Relative changes in mean square charge radii for the mercury isotopes. The dots represent the data for the ground state and the triangles are the results for the isomeric state. The solid line is the prediction for the spherical droplet model, normalized to ^{198}Hg . The data is taken from [14] [15].

The mean square charge radius of the mercury isotopes exhibits a large odd-even staggering for the neutron-deficient mercury isotopes (figure 1.9), which is suggested to be a manifestation of shape coexistence [9]. Shape coexistence is the phenomenon in which two (or more) distinct types of deformation occur at low energy in one and the same nucleus. Throughout the years, the status of shape coexistence has evolved from an exotic rarity which exhibits “islands of occurrence” to the current position in which it occurs in several different regions of the nuclear chart [9]. Shape coexistence has not yet been observed in all nuclei. The most extensive manifestation of shape coexistence is situated in the neutron-deficient isotopes near $Z = 82$ [16]. Besides mercury, another intriguing example

of shape coexistence in this region is ^{186}Pb . The ground state and first two excited states of ^{186}Pb are 0^+ states with an assumed transition in shape from spherical for the ground state towards oblate for the first excited state and prolate for the second excited state. This is schematically illustrated in figure 1.10.

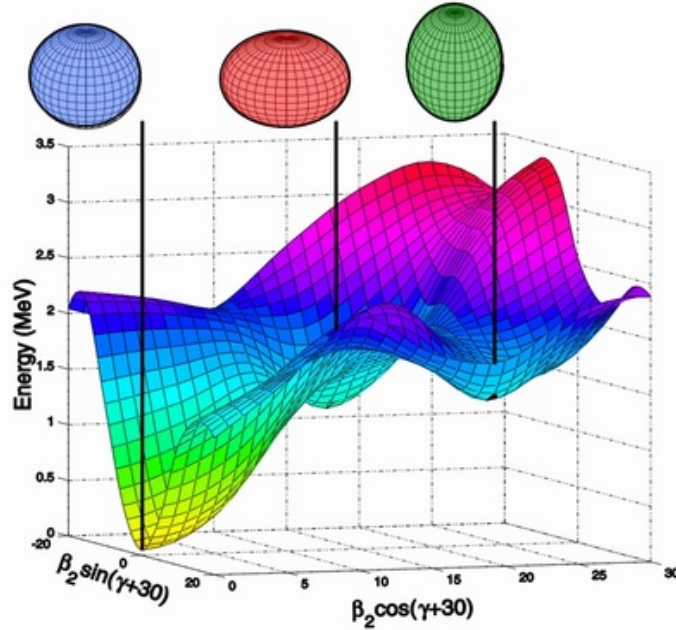


Figure 1.10: Theoretical calculation for the energy surface of ^{186}Pb , which shows the three different shapes (spherical, oblate and prolate) for the ground state and the first two excited states [17].

Understanding the occurrence of shape coexistence in atomic nuclei is one of the greatest challenges faced by theories of nuclear structure. The investigation of the neutron-deficient mercury isotopes ($Z = 80$) plays an important role in the understanding of shape coexistence in this region of the nuclear chart. The ground state of $^{186-192}\text{Hg}$ and the even-even $^{182,184}\text{Hg}$ isotopes is a slightly oblate deformed, while the ground state of the odd $^{181,183,185}\text{Hg}$ isotopes is strongly prolate deformed. One believes [18] that this can be explained by the presence of a highly deformed intruder state which lowers in energy in the more neutron-deficient isotopes and reaches a minimum around mid-shell ($N = 104$). (In an intruder state particles are excited across a shell gap.) This intruder state becomes the ground state for the odd $^{181,183,185}\text{Hg}$ isotopes. One expects, based on shell model calculations [18] (discussed in more detail in subsection 1.4.2), that the ground state of the odd mercury isotopes returns to a slightly deformed oblate state due to the rise of the energy level of the intruder state for neutron-deficient isotopes with mass below ^{181}Hg . In a recent laser-spectroscopy experiment performed at the ISOLDE facility in CERN, the hyperfine spectra were measured for 15 different mercury isotopes ranging from ^{177}Hg to ^{208}Hg . One of the main goals of this experiment was to extend the currently existing knowledge of the mean square charge radius behavior down towards the more neutron-deficient isotopes and verify if the mercury isotopes become spherical again or whether the more neutron-deficient nuclei become even more deformed. In this thesis work, a subset of the data obtained in this experiment was analyzed in order to verify the consistency between the old measurements and the new approach of in-source laser ionization spectroscopy used in this work.

In what follows, a short historical overview of the measurements performed on the mercury isotopic chain are given. The chapter ends with a discussion of the theoretical predictions of the shell model calculations.

1.4.1 Experimental measurements of the charge radii in the mercury isotopes

Historical overview

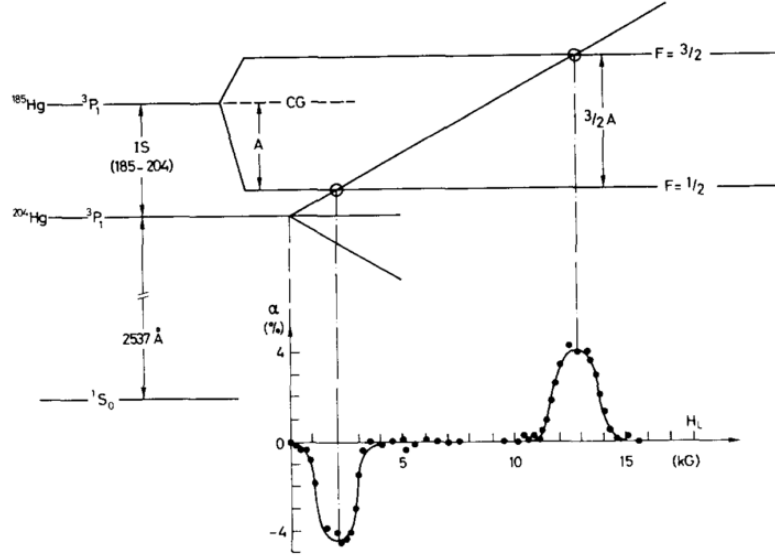


Figure 1.11: In the lower part of the graph the experimental β -asymmetry of ^{185}Hg is shown as a function of the scanning magnetic field. In the upper part the deduced hyperfine splitting of the 3P_1 state of ^{185}Hg is shown. From the position of the center of gravity the isotope shift can be determined relative to ^{204}Hg and from this the relative change in mean square charge radii $\delta \langle r^2 \rangle$ [19].

By performing optical spectroscopy studies on isotopic chains, one can gain information on the ground state and isomeric state of the nucleus under consideration. Using optical spectroscopy, properties such as the hyperfine splitting and isotope (isomer) shift of ground states and isomers can be obtained. From these measurements the spin I , the magnetic dipole moment μ , the spectroscopic quadrupole moment Q_s and the change in mean square charge radius $\delta \langle r^2 \rangle$ can be determined. This is explained in chapter 2. First measurements of the isotope shifts of mercury were performed in 1963 [20] and 1966 [21] using conventional optical spectroscopy in the isotope range $192 \leq A \leq 204$. An extension of the measurements on the odd isotopes of mercury with $181 \leq A \leq 191$ were done in 1971 using β -radiation detected optical pumping method (β -RADOP) [19]. The basic principles of the (β -RADOP) technique which was used in [19] is shown in figure 1.11.

A ^{204}Hg light source was placed in a magnetic field, causing the spectral line to be split into three components due to Zeeman splitting. When a Zeeman component of ^{204}Hg matches with a hyperfine component of the unstable isotope, ^{185}Hg in the case of the illustration, the nuclear spin is polarized by means of optical pumping. This polarization was detected via the angular asymmetry of the β -decay.

Because only two resonant absorptions were detected, the nuclear spin I of ^{185}Hg is $1/2$. From the sign difference of the β -decay asymmetry measured either in vacuum or in the presence of a buffer gas, it is possible to say which absorption corresponds to the $^1S_0 - F = 1/2$ or $^1S_0 - F = 3/2$ transition in the unstable mercury isotope. From this information, the center of gravity CG, the isotope shift IS, magnetic dipole moment μ and the change in mean square charge radius $\delta \langle r^2 \rangle$ could be deduced.

This measurement showed the surprising result of a large change in the $\delta \langle r^2 \rangle$ between ^{187}Hg and ^{185}Hg . Calculations based on the Strutinsky shell-correction method [13] interpreted this result as a transition from a slightly deformed oblate state for the heavier isotopes towards a strongly deformed prolate state for the lighter isotopes (see figure 1.8). Even isotopes in the mass range $182 \leq A \leq 190$ were measured with (collinear) laser spectroscopy [14], whose sensitivity had been improved during the years. Until recently, the isotope shifts and hyperfine splitting of mercury had been measured from ^{181}Hg to ^{206}Hg .

Experimental energy levels for even-even mercury isotopes

In order to get a full picture of shape coexistence and nuclear structure in this region of the nuclear chart, different experimental techniques have to be applied. As such, not only isotope (isomer) shifts/hyperfine splitting add to the discussion of shape coexistence, but observables as for instance energy level structures or $B(E2)$ values obtained from in-beam gamma spectroscopy or Coulomb-excitation experiments are of help. These experiments are added to obtain the picture shown in figure 1.12. Here, one observes two rotational bands built on a 0^+ level in the even-even mercury isotopes, where the excitation energy of the excited band behaves parabolic with respect to neutron number and reaches a minimum around mid-shell. All these experimental values can be compared to state-of-the-art nuclear theory such as e.g. in [18].

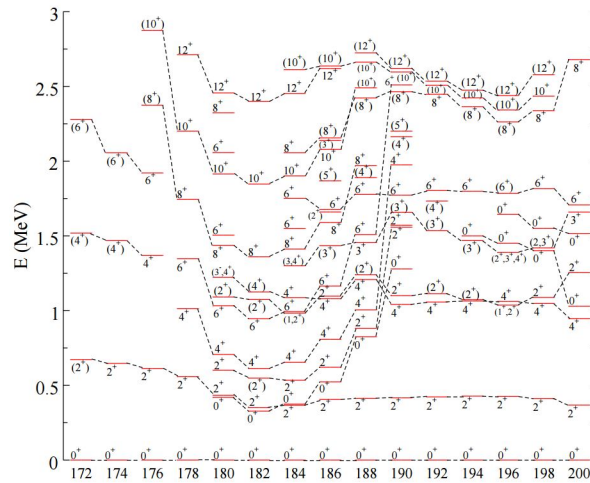


Figure 1.12: Systematics of excited states in the even-even mercury isotopes. Only levels up to an excitation energy of 3 MeV are shown. Note the presence of a rotational band built on an excited 0^+ state with a “parabolic-like” energy dependence centered around $N = 102$ [18].

1.4.2 Theoretical considerations

This section gives a short overview of theoretical approaches used to describe shape coexistence with a focus on a study of the even-even mercury isotopes. This section is based on [9][18], where more information can be found.

The origin of the appearance of shape coexistence at a certain neutron and proton number can be found in the balance between two opposing tendencies: on one side the stabilizing effect of closed shells and sub-shells which prefers the nucleus to have a spherical shape, versus the residual interactions between protons and neutrons which like to redistribute the nucleons in a more deformed shape. Nowadays mainly two approaches are used to understand the appearance of shape coexistence: microscopic shell model descriptions and mean field descriptions.

In the shell model approach, multiple particle-hole (mp-nh) pair excitations across “closed” shells can give rise to low-lying shape coexistence configurations. In some cases the deformed (mp-nh) configuration has a lower energy than the (0p-0h) spherical state, such as in the island of inversion. The energy level of the (mp-nh) configuration is determined by the balance between the cost in energy to excite nucleon-pairs across a “closed” shell and the energy gain due to more protons and neutrons available to interact through the residual interactions, referred to as the correlation energy gain. The proton-neutron interaction energy, which is mainly multiplicative in the number of interacting neutron times the number of interacting protons, has a major contribution in the correlation energy gain. The (mp-nh) configuration becomes a low-lying state if the reduction of the original spherical shell gap due to the monopole shifts and the correlation energy gain are large enough to make the intruder state energetically favorable.

The mean field approach uses self-consistent Hartree-Fock theory. The Hartree-Fock energy is minimized while constraining the nuclear density distribution to exhibit specific values for the quadrupole deformation parameter β . These calculations result in an energy surface with the minimal energy which can be situated in a deformed configuration. In order to compare the results of mean field calculations with experimental data, all broken symmetries have to be restored to construct states with a correct spin I (and projection K). These states form the basis to calculate collective properties (i.a. charge radii, quadrupole moments, energy spectra) going beyond the mean field approach.

A study of even-even mercury isotopes

In the case of mercury ($Z = 80$) the model space includes the regular proton $2h$ configuration and a number of neutrons outside of the $N = 82$ closed shell which are allowed to move inside the open neutron shell $N = 82 - 126$ as well as the proton $2p-4h$ (a proton pair is excited across the $Z = 82$ shell gap) configuration and the same number of neutrons. Considering all possible partitions of the active neutrons and protons over this large model space, is far beyond present computational possibilities. Therefore the model space has to be restricted using the interacting boson model approximation (IBM). This approach only considers nucleon pairs coupled to $J = 0$ and $J = 2$ which are treated as bosons.

The energy of the lowest-lying intruder 0^+ state $E_{\text{intruder}}(0^+)$ is equal to

$$E_{\text{intruder}}(0^+) = 2(\epsilon_{j_\pi} - \epsilon'_{j'_\pi}) - \Delta E_{\text{pair}} + \Delta E_{\text{M}} + \Delta E_{\text{Q}} \quad (1.16)$$

with the first term describing the energy needed to excite 2 protons across the unperturbed $Z = 82$ shell gap, the second term describes the energy gain when forming two extra 0^+ coupled pairs (particle and hole pairs), the third term describes the change of the single-particle gap $Z = 82$ as a function of the neutron number and the last term is equal to the quadrupole energy gain when opening up the proton shell, relative to the regular configuration. Figure 1.13 shows the contribution of the different energy terms to the excitation energy of the lowest-lying 0^+ intruder state in lead, which is a proton $2p$ - $2h$ configuration. The balance in cost and gain in energy results in a “parabolic-like” behavior of the excitation energy with a minimum around mid-shell ($N = 104$). A similar energy consideration for the lowest-lying 0^+ intruder state in mercury, which is the $2p$ - $4h$ configuration, results in an analogue energy dependence on neutron number. This is depicted in figure 1.14a and is also visible in the experimental energy levels (figure 1.12).

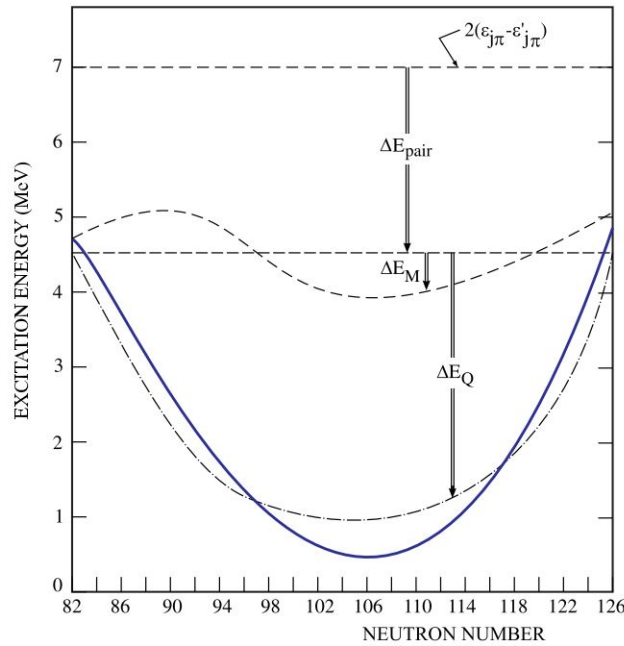


Figure 1.13: The different energy terms contribution to the excitation energy of the lowest-lying $2p$ - $2h$ 0^+ intruder state in the lead isotopes. The quadrupole energy gain ΔE_Q , the monopole energy shift ΔE_M , the pairing energy gain ΔE_{pair} and the unperturbed energy $2(\epsilon_{j\pi} - \epsilon'_{j\pi})$ are displayed in a schematic way [9].

When two states with the same spin I and parity come close in energy, mixing between these states occurs. In the mercury isotopes the ground state and intruder state (with the same spin I and parity) come close in energy (figure 1.14a) and configuration mixing takes place. The theoretical energy levels obtained with interacting boson model with configuration mixing (IBM-CM) are displayed in figure 1.14b. The regular content in the wave function of the two lowest-lying 0^+ , 2^+ , 3^+ and 4^+ states is shown in figure 1.14c. A textbook example of two-state mixing is visible if one compares the excitation energy of the regular and intruder 2^+ state in figure 1.14a with figure 1.14b. Without including configuration mixing, the two 2^+ states cross. If one includes configuration mixing, the two states will start “repelling” each other near the crossing point and interchange identity as visible in figure 1.14c. The 0^+ regular state and intruder state come close in energy

around mid-shell. This results in an increasing component of the intruder state in the ground state starting from $A = 192$ with a maximum of 15% around $A = 182, 184$. This mixture with a deformed state is visible in the mean square charge radius $\langle r^2 \rangle$ which is a linear combination of $\langle r^2 \rangle_{\text{regular}}$ and $\langle r^2 \rangle_{\text{intruder}}$. Studies of the experimental $\langle r^2 \rangle$ for the ground state indicate a maximum admixture of 20% in the interval $180 \leq A \leq 200$. A comparison between the theoretical and experimental changes in mean square charge radius is displayed in figure 1.15. Theory and experiment agree. The deviation from the spherical droplet model line (the dotted line) can be explained by the effect of a slightly more deformed regular configuration when opening the neutron shell and by an increased deformed regular component starting from $A = 192$.

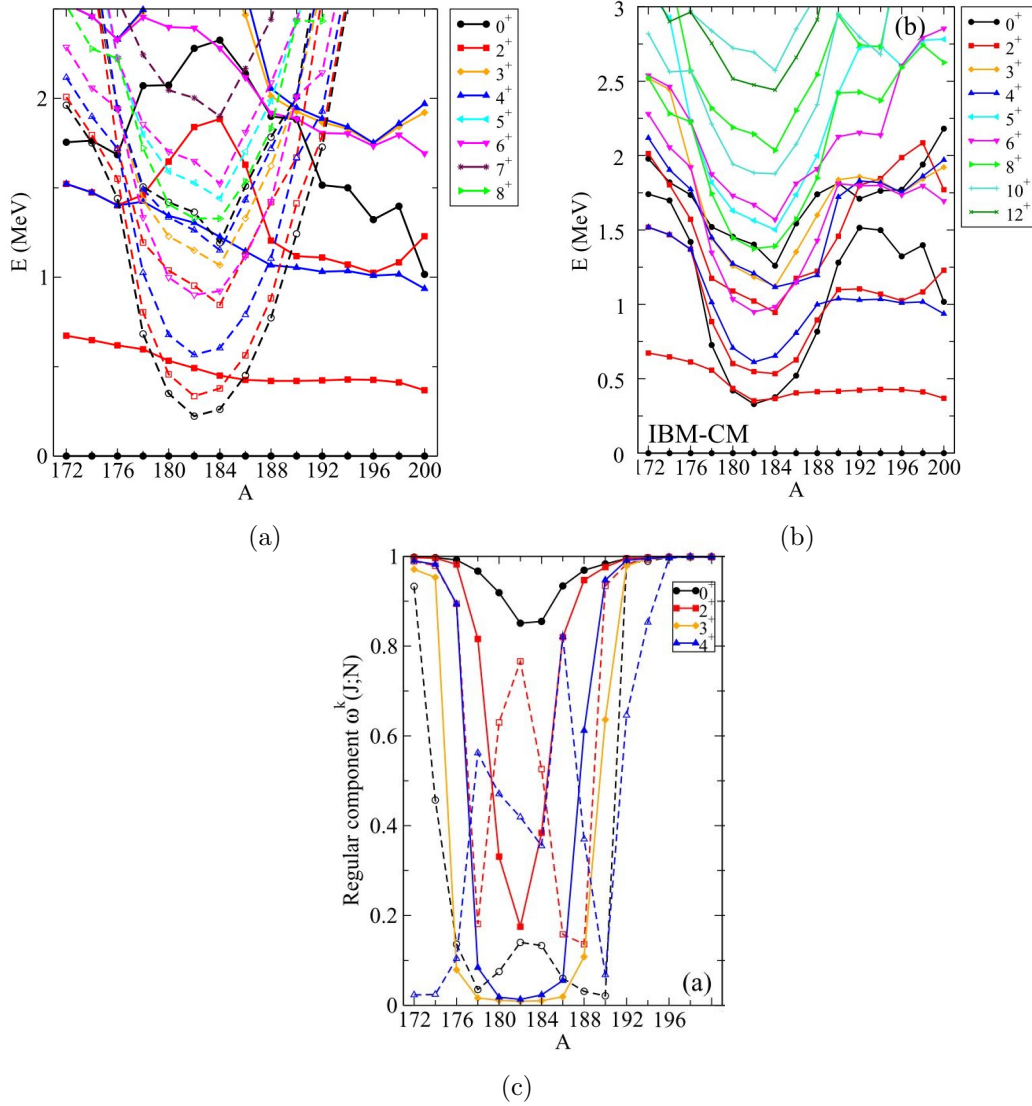


Figure 1.14: Results from IBM-CM calculations for the even-even mercury isotopes [18]. (a) Energy spectra without including configuration mixing. The two lowest-lying regular states (full lines with closed symbols) and intruder states (dashed lines with open symbols) for each of the total angular momenta are displayed. (b) Theoretical obtained energy levels with IBM including configuration mixing. (c) Regular content of the two lowest-lying states for each value of the total angular momenta. First state: closed lines with closed symbols. Second state: dashed lines with open symbols.

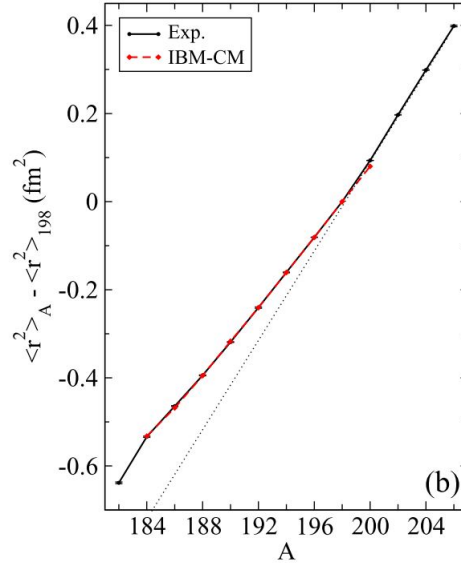


Figure 1.15: Comparison between theoretical and experimental values for the change in mean square charge radius $\langle r^2 \rangle$ for the even-even mercury isotopes. Dashed line: spherical droplet model behavior [18].

Odd-mass mercury isotopes

The IBM description is fundamentally limited to the even-even cases, so while it is very useful to understand the basic mechanism driving the lowering of the intruder state at mid-shell, it does not describe the difference between odd and even isotopes. However, those calculations give a good starting point to understand why the intruder state becomes the ground state around mid-shell. One can learn from figure 1.14a that around mid-shell the intruder state comes close in energy to the regular (ground) state. The addition of an odd neutron will lower the energy of the intruder state in such a way that this deformed state becomes the ground state in $^{181,183,185}\text{Hg}$. This can intuitively be understood by observing the Nilsson diagram (figure 1.7). According to the spin and parity of ^{187}Hg ($I^\pi = 3/2^-$) the odd neutron occupies the $3p_{3/2}$ level in the spherical state, while according to experiment the odd neutron most probably occupies the $1/2 [521]$ Nilsson level in the deformed state. This Nilsson level is the one into which the $3p_{3/2}$ transforms if the nucleus deforms. Due to the repulsion from the $1/2 [510]$ level, the $1/2 [521]$ level strongly decreases in energy with increasing deformation. The odd neutron can thus significantly lower its energy by coupling to a deformed nucleus. Therefore the energy of the deformed state will lower more compared to the intruder state in the even-even mercury isotopes and it even becomes the ground state around mid-shell. Of course theory is still trying to describe these effects quantitatively (odd isotopes are exceptionally difficult to calculate), but the basic mechanism can be described using this combination of different approaches.

Chapter 2

Laser spectroscopy

Laser spectroscopy refers to measuring atomic transitions with a high precision by applying excitations/ionizations induced by laser light. Laser ionization spectroscopy is used to increase the sensitivity (ions can be easily manipulated with electric and magnetic fields). An example of a laser ionization scheme is displayed in figure 2.1.

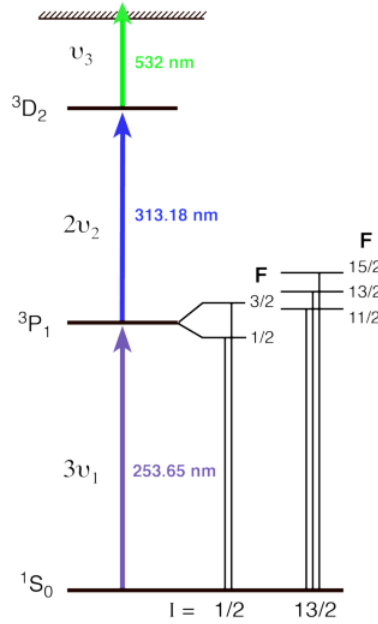


Figure 2.1: A three-steps laser ionization scheme [15]. Left: Atomic transitions. Right: Hyperfine splitting indicating the nuclear spin I and the total spin F .

The atomic energy levels depend on nuclear properties. Therefore one can gain a lot of information on the ground and isomeric state of a nucleus by performing laser spectroscopy studies. This technique allows one to measure the hyperfine splitting and the isotope (isomer) shift. From these measurements the spin I , the magnetic dipole moment μ , the spectroscopic quadrupole moments Q_s and the change in mean square charge radii $\delta \langle r^2 \rangle$ can be determined.

In this chapter a theoretical discussion of the isotope (isomer) shift, fine structure and hyperfine structure is presented.

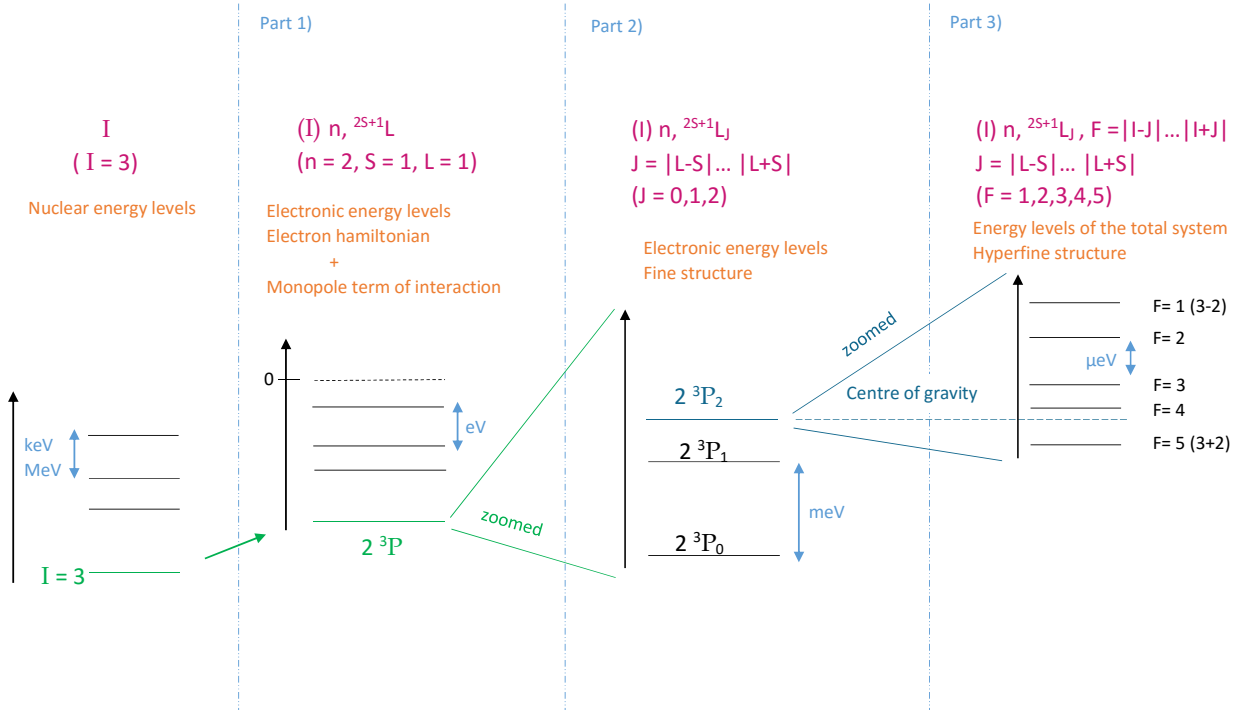


Figure 2.2: Outline of how the energy levels of an atom are determined [22]. The total orbital angular momentum L , the total spin S and the total angular momentum J of the electrons is indicated together with the nuclear spin I and the total spin of the atom F .

2.1 Isotope (isomer) shift

In this section, the isotope (isomer) shift will be explained. Their physical origin will be discussed together with the information that can be obtained by measuring the isotope (isomer) shift.

In figure 2.2 an outline of the determination of atomic energy levels is shown. The energy levels in part 1 of the figure are isotope (isomer) dependent. The isotope shift $\delta v^{A,A'}$ is defined as $v^{A'} - v^A$ with $v^{A'}$ and v^A the frequency of the electromagnetic radiation to excite the isotope (or isomer) with mass number A' and A respectively from one energy level in part 1 to an other energy level in this part. Since the fine structure is isotope (isomer) independent, the isotope (isomer) shift can also be determined by the difference in frequency for an excitation from one energy level in part 2 to another. When the experimental resolution is good enough to observe the hyperfine splitting, the center of gravity has to be determined in order to be able to determine the isotope shift. More information about the fine and hyperfine structure is given in section 2.2.

The energy levels in part 1 of figure 2.2 change from one isotope (or isomer) to another because of the change in mass and size of the nucleus. These contributions to the isotope shift $\delta v^{A,A'}$ are called the mass shift $\delta v_{MS}^{A,A'}$ and field shift $\delta v_{FS}^{A,A'}$, respectively.

2.1.1 Mass shift

When the energy levels in part 1 of figure 2.2 are calculated, the nucleus is assumed to be infinitely heavy and considered to be a point charge. In this approximation the center of mass of the atom coincides with the center of the nucleus and the center of the inertial coordinate system can be put in the center of the nucleus. The Hamiltonian of such a system consists of the kinetic energy of the electrons, the electron-electron interaction and the Coulomb interaction between the electrons and the nucleus. When the electron-electron interaction is replaced by the interaction of an electron with a spherical symmetric potential, the Hamiltonian is rotational invariant. The energy levels are characterized by the quantum numbers L, S and there is an degeneration according to m_l and m_s . A point to notice is that the energy levels depend on the electron mass, when the electron mass would be higher the electrons would be less bound. Because for one electron the Coulomb interaction with all isotopes (isomers) is the same, the calculated energy levels in this approximation are isotope (isomer) independent.

The mass shift originates from the fact that the nucleus is not infinitely heavy. This causes the electrons and the nucleus to move around their common center of mass and the motion of the nucleus has to be included in the Hamiltonian. It is shown [23] that this new Hamiltonian can be written as the sum of a Hamiltonian describing the center of mass motion and a Hamiltonian that describes the relative movement. The latter looks very similar to the Hamiltonian which only considers the electrons to move around the nucleus. So taking into account the motion of the nucleus will not lift any degeneration, but will only shift the energy levels. The energy levels shift because of the differences between the Hamiltonian where the nucleus is infinitely heavy and the total Hamiltonian which includes the nucleus moving around the common center of mass. These differences lead to the so-called normal mass shift $\delta v_{NMS}^{A,A'}$ and the specific mass shift $\delta v_{SMS}^{A,A'}$.

The origin of the normal mass shift is the fact that the electron mass has to be replaced with the reduced mass in the part of the Hamiltonian that describes the kinetic energy of the electrons. Because the reduced mass is larger than the electron mass, the electrons will be less bound. The normal mass shift can be written as

$$\delta v_{NMS}^{A,A'} = \frac{m_{A'} - m_A}{m_A \cdot m_{A'}} \cdot m_e v \quad (2.1)$$

with m_e the mass of the electron, $m_A, m_{A'}$ the mass of the isotope (isomer) with mass number A and A' respectively and v the frequency of the transition.

An extra term appears in the Hamiltonian that describes the relative motion when it is compared to the Hamiltonian where the nucleus is considered infinitely heavy. This term consists of a correlation between the electronic momenta and depends on the mass of the nucleus. This extra term is the origin of the specific mass shift. The specific mass shift can be written as

$$\delta v_{SMS}^{A,A'} = \frac{m_{A'} - m_A}{m_A \cdot m_{A'}} \cdot K_{SMS} \quad (2.2)$$

where K_{SMS} is a constant that depends on the electronic transition, but which is isotope (isomer) independent. The $^1S_0 \rightarrow ^3P_1$ transition used in the mercury experiment has a specific mass shift constant $K_{SMS} = (0 \pm 0.5)m_e v$, with $m_e v$ the normal mass shift constant [14]. The mass shift contribution to the isotope shift is smaller, when the mass of the nucleus is larger. This can easily be understood because when the mass of the nucleus is bigger, the center of mass coincides more with the center of the nucleus.

Typical orders of magnitude for the mass shift are between GHz (light elements, $Z \approx 10$) and 10MHz (heavy elements, $Z \approx 80$) per atomic mass unit [24].

2.1.2 Field shift

When the nucleus is no longer considered a point-like particle by taking into account the nuclear volume, the energy levels in part 1 of figure 2.2 shift. This shift of the energy levels is proportional to the electronic density at the origin $\rho_e(O)$ and the mean square charge radius $\langle r^2 \rangle$ of the nucleus. This can intuitively be explained. When the nucleus would be a point particle the electric potential at the origin goes to infinite. But when the nucleus is spherical and has a certain volume, the electric potential has finite values inside the nucleus. So when the nucleus has a certain volume, the total energy of electrons who can penetrate the nucleus will be smaller than when the nucleus would be a point charge. In figure 2.3 an illustration is given of the Coulomb potential for different charge radii of the nucleus.

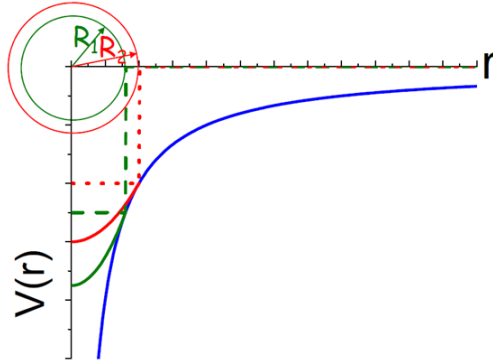


Figure 2.3: Coulomb potential for different charge radii of the nucleus. The blue line corresponds to the Coulomb potential for a point charge nucleus [25].

This contribution to the isotope shift is called the field shift. Typical orders of magnitude for the field shift are between 10 MHz (light elements, $Z \approx 10$) and 10 GHz (heavy elements, $Z \approx 80$) [24] per atomic mass unit. The field shift contribution to the isotope (isomer) shift is given by

$$\delta v_{FS}^{A,A'} = -\frac{Ze^2}{6\epsilon_0 h} \Delta |\Psi(0)|^2 \lambda^{AA'} = F^{AA'} \lambda^{AA'}. \quad (2.3)$$

The F-factor $F^{AA'}$, also called the field shift constant, is a parameter that depends on the electronic transition, through the difference between the electronic density at the origin between the upper and lower electronic level $\Delta |\Psi(0)|^2$. The F-factor has a slight dependence [26] on the mass number which is given by

$$F^{AA'} = ((A + A')/(A'' + A'''))^{-0.125} F^{A''A'''}. \quad (2.4)$$

The $^1S_0 \rightarrow ^3P_1$ transition used in the mercury experiments has a F-factor $F^{198\ 204} = -55.36$ GHz/fm² with an assumed uncertainty of 7%. This value is based on ab initio MCDF calculations [14].

Parameters	$A' = 181$	$A' = 182$
$\frac{\delta \langle r^4 \rangle^{198 A'}}{\delta \langle r^2 \rangle^{198 A'}}$	68	68
$\frac{\delta \langle r^6 \rangle^{198 A'}}{\delta \langle r^2 \rangle^{198 A'}}$	3761	3774
C_1	1570	
C_2	-1.71	
C_3	0.00455	

Table 2.1: C parameters for the mercury isotopes [27] and the ratio of the different orders $\delta \langle r^4 \rangle^{198 A'}$ and $\delta \langle r^6 \rangle^{198 A'}$ to the changes in the mean $\delta \langle r^2 \rangle^{198 A'}$ for $A' = 181$ or 182 calculated by the liquid drop model taking $A = 198$ as a reference.

The nuclear parameter $\lambda^{AA'}$ depends on the change of the nuclear charge distribution between isotopes (isomers) with atomic mass number A' and A . It can be written as

$$\lambda^{AA'} = \delta \langle r^2 \rangle^{AA'} \left(1 + \frac{C_2 \delta \langle r^4 \rangle^{AA'}}{C_1 \delta \langle r^2 \rangle^{AA'}} + \frac{C_3 \delta \langle r^6 \rangle^{AA'}}{C_1 \delta \langle r^2 \rangle^{AA'}} + \dots \right) \quad (2.5)$$

with $\delta \langle r^2 \rangle^{AA'} = \langle r^2 \rangle^{A'} - \langle r^2 \rangle^A$ and the coefficients $C_1, C_2, C_3, \dots$ have been calculated by Seltzer [27]. Under the assumption that the nucleus is spherically symmetric with a constant nuclear density and radius $R = R_0 A^{1/3} = 1.2 A^{1/3}$ fm, $\langle r^2 \rangle^A = 3/5 R_0^2 A^{2/3}$, $\langle r^4 \rangle^A = 3/7 R_0^4 A^{4/3}$ and $\langle r^6 \rangle^A = 1/3 R_0^6 A^2$. The parameters C_i and the ratio of the different orders to the changes in the mean square charge radius for $^{181,182}\text{Hg}$ are given in table 2.1. The relation in equation 2.5 for $^{181,182}\text{Hg}$ with reference ^{198}Hg becomes

$$\lambda^{198 \ 181,182} = 0.937 \delta \langle r^2 \rangle^{198 \ 181,182} . \quad (2.6)$$

A more recent and reliable approach determines the higher radial moments (higher than $\langle r^2 \rangle$) by combining experimental data from muonic atom spectroscopy and elastic electron scattering measurements. The contribution of the higher radial moments to $\lambda^{AA'}$ averaged over different isotopes is equal to -7.3% for mercury [28]. This results in

$$\lambda^{198 \ 181,182} = 0.927 \delta \langle r^2 \rangle^{198 \ 181,182} . \quad (2.7)$$

Equation 2.5 denotes that the change in the mean square charge radius $\delta \langle r^2 \rangle^{AA'}$ can be determined by measuring the isotope shift, using knowledge of the F- and K_{SMS} -factors. But in order to be sensitive to the field shift a transition between levels with a different number of s -or $p_{1/2}$ -electrons has to be measured.

2.1.3 King plot

Combining the mass shift and the field shift, the isotope (isomer) shift becomes

$$\delta v^{A,A'} = \frac{m_{A'} - m_A}{m_A \cdot m_{A'}} \cdot (m_e v + K_{SMS}) + F \cdot \lambda^{AA'} = M \frac{m_{A'} - m_A}{m_A \cdot m_{A'}} + F \cdot \lambda^{AA'} \quad (2.8)$$

with M the mass-shift constant. The K_{SMS} constant depends on the electronic transition and is isotope (isomer) independent. The F -factor also depends on the atomic transition, but has a slight dependence on the mass number. These constants can be calculated by atomic theory. If these calculations are not available, the values can be deduced from a so-called King plot if the atomic factors of another transition of which the isotope shifts were measured with another technique are known. In a King plot the modified isotope shifts $\mu\delta v_2^{AA'} = \frac{m_A \cdot m_{A'}}{m_{A'} - m_A} \delta v_2^{AA'}$ of the transition with known constants are plotted as a function of the modified isotope shifts $\mu\delta v_1^{AA'} = \frac{m_A \cdot m_{A'}}{m_{A'} - m_A} \delta v_1^{AA'}$ of the transition with unknown constants. From the linear relationship between the modified isotope shifts, given by

$$\mu\delta v_2^{AA'} = \frac{F_2^{AA'}}{F_1^{AA'}} \mu\delta v_1^{AA'} + M_2 - \frac{F_2^{AA'}}{F_1^{AA'}} M_1 \quad (2.9)$$

with $F_2^{AA'}/F_1^{AA'}$ a constant factor, the unknown $F_1^{AA'}$ and M_1 constants can be deduced from the slope and the intercept with the y-axis of the line formed by the graph of $\mu\delta v_2^{AA'}$ against $\mu\delta v_1^{AA'}$, respectively.

2.1.4 Changes in mean square charge radii and deformation

As explained in section 2.1.2, the difference in mean square charge radii can be determined using optical-spectroscopy techniques. To obtain an absolute value of the $\langle r^2 \rangle$, an absolute measurement for the reference isotope together with a measurement of the difference in mean square charge radius $\delta \langle r^2 \rangle^{AA'}$ between the isotope under investigation (A') and the reference isotope (A) is necessary. Elastic electron scattering for instance, is such a method which provides an absolute value for $\langle r^2 \rangle$. Because of the need for targets, so far only stable isotopes could be investigated with this technique.

There are two contributions to the change $\delta \langle r^2 \rangle^{AA'}$. When more nucleons are added to the nucleus the volume changes and this induces a change in $\langle r^2 \rangle$ (denoted as $\delta \langle r^2 \rangle_{\text{spherical}}^{AA'}$), but also when the shape of the nucleus changes at a constant density $\langle r^2 \rangle$ will change (denoted as $\delta \langle r^2 \rangle_{\text{deformation}}^{AA'}$). This leads to

$$\delta \langle r^2 \rangle^{AA'} = \delta \langle r^2 \rangle_{\text{spherical}}^{AA'} + \delta \langle r^2 \rangle_{\text{deformation}}^{AA'} \quad (2.10)$$

It is shown [24] that this can be parametrized as

$$\delta \langle r^2 \rangle^{AA'} = \delta \langle r^2 \rangle_{\text{spherical}}^{AA'} + \frac{5}{4\pi} \overline{\langle r^2 \rangle_{\text{spherical}}} \delta \langle \beta^2 \rangle^{AA'} \quad (2.11)$$

in which $\overline{\langle r^2 \rangle_{\text{spherical}}} = (\langle r^2 \rangle_{\text{spherical}}^A + \langle r^2 \rangle_{\text{spherical}}^{A'})/2$ and β is the quadrupole deformation parameter that is nuclear model dependent. The values $\delta \langle r^2 \rangle_{\text{spherical}}^{AA'}$ and $\langle r^2 \rangle_{\text{spherical}}$ are taken from the droplet model. Equation 2.11 denotes that $\delta \langle \beta^2 \rangle^{AA'}$ can be determined from $\delta \langle r^2 \rangle^{AA'}$. A value of $\langle \beta^2 \rangle$ can be determined if $\langle \beta^2 \rangle$ is known for at least one isotope. A value of $\langle \beta^2 \rangle$ was deduced for some even mercury isotopes from the transition probability $B(E2)$ [14]. It is important to notice that there is no sensitivity to the sign of β . So from this it can not be concluded if the nucleus is oblate ($\beta < 0$) or prolate ($\beta > 0$) deformed.

2.2 Fine and hyperfine structure

In the next two sections the basic principles of the fine and hyperfine structure are explained. A complete description can be found in [22].

2.2.1 Fine structure

Up to now, relativistic effects were neglected. If relativistic effects are included, the Schrödinger equation has to be replaced by the Dirac equation. Up to first order in $(v/c)^2$ the Dirac equation equals the Schrödinger equation plus three correction terms. The first correction is the mass-velocity effect. Electrons close to heavy nuclei have relativistic velocities and this leads to a small shift of the energy levels with quantum numbers (nLS) with L the orbital angular momentum and S the spin. Such a correction is called a scalar correction. Also the mass effect and volume effect, explained in section 2.1.1 and 2.1.2 respectively, are scalar corrections. The second correction is the Darwin effect. The electrons move very fast on a very short time scale and feel a smeared out Coulomb potential. The mass-velocity effect and Darwin effect give isotope independent scalar corrections. This in contrast to the mass effect and volume effect.

The third correction is the spin-orbit coupling. When the electron cloud has orbital angular momentum, they create a magnetic field. This magnetic field interacts with the spin magnetic moment of the electrons and gives a correction term to the Hamiltonian that is proportional to $\hat{L} \cdot \hat{S}$. In the calculation of the energy levels in part 1 of figure 2.2 this term is not included and the energy levels are degenerate according to the total angular momentum J with $J = |L - S| \dots |L + S|$. The energy levels are independent of the orientation of L with respect to S . But when relativistic effects are included, this degeneration is lifted because of the electronic energy dependence on the orientation of their orbital angular momentum with respect to their spin. The energy levels are now characterized by the quantum numbers (L, S, J) . The ground state level can be found by applying Hund's rules. An example is shown in part 2 of figure 2.2.

2.2.2 Hyperfine structure

Up until now only the Coulomb interaction between a spherical nucleus and the electron cloud is discussed. But there is also a magnetic interaction between the nucleus and the electrons. The moving protons inside the nucleus create a convection current and the spin of the neutrons and protons create spin currents. These currents interact with the current created by the electrons in the electron cloud. This leads to the so-called magnetic dipole splitting of the energy levels.

Moreover, a spherically symmetric charge distribution of the nucleus was assumed. In reality the nucleus can be deformed and the energy will then depend on the orientation of J , because the rotational symmetry is broken. This leads to the so-called quadrupole splitting of the energy levels.

Taking into account the hyperfine structure results in part 3 of figure 2.2.

Magnetic dipole moment

The electron cloud creates a magnetic field at the position of the nucleus that is proportional to the total angular momentum J . The magnetic dipole moment of the nucleus,

which is proportional to the nuclear spin I , interacts with this magnetic field. This interaction adds a perturbation term, which is proportional to $\hat{I} \cdot \hat{J}$, to the Hamiltonian. Before, the energy levels were described by the quantum numbers ($LSJI$) and the energy was independent of the orientation of the nuclear spin I with respect to J . The magnetic dipole interaction lifts this degeneracy. The energy levels are now characterized by the quantum number F and are degenerated with respect to F_z . The shift in energy is given by

$$\Delta E_A = -\frac{A}{2}C \quad (2.12)$$

with $C = F(F+1) - I(I+1) - J(J+1)$ and $A = \frac{\mu B_e(0)}{IJ} = \frac{\mu_n g_I B_e(0)}{J}$. μ is the magnetic dipole moment, g_I the g-factor and $B_e(0)$ the magnetic field created by the electron cloud at the center of the nucleus. $B_e(0)$ depends on the electron configuration. The magnetic dipole interaction leads to a non equidistant energy splitting.

Since $B_e(0)$ is isotope independent, the g-factor of the nucleus can be measured when the A-factor and g-factor of an other isotope are known.

Quadrupole moment

When the Hamiltonian that describes the Coulomb interaction between the nucleus and the electron cloud is expanded in a Taylor series, the first term is the monopole term. The monopole term describes the Coulomb interaction between the electron cloud and a point-like nucleus with charge Z . The second term is the dipole term and vanishes because of the negative parity of the dipole operator. The third term is the quadrupole term and is responsible for the quadrupole splitting of the energy levels.

In part 2 of figure 2.2 the energy levels of the atom are described by the quantum numbers ($LSJI$). Because there is no interaction between the total angular momentum of the electron cloud J and the spin of the nucleus I , the energy is the same for all orientation of the nuclear spin I with respect to the angular momentum of the electron cloud J and the energy levels are degenerated. The quadrupole term of the Coulomb interaction can be seen as a perturbation term. This perturbation will lift this degeneracy and the energy depends on the orientation of the nuclear spin I with respect to the angular momentum of the electron cloud J . The good quantum numbers are now (JIF) with $F = |J - I| \dots |J + I|$. Because the total rotation of J and I simultaneous doesn't change the energy, there will be a degeneracy with respect to the projection of F , namely F_z . The shift in energy levels is given by

$$\Delta E_B = B \frac{3C(C+1) - 4I(I+1)J(J+1)}{8(2I-1)I(2J-1)J} \quad (2.13)$$

with $C = F(F+1) - I(I+1) - J(J+1)$ and $B = eQ_s \frac{\partial^2 V}{\partial z^2}$. The spectroscopic quadrupole moment Q_s is zero for $I = 0, 1/2$ and the electric field gradient produced by electrons $\frac{\partial^2 V}{\partial z^2}$ with $J = 0, 1/2$ is also zero. This can be shown by applying the Wigner-Eckart theorem. The spectroscopic quadrupole moment is defined as

$$Q_s = Z2\sqrt{\frac{4\pi}{5}} \langle I, I | r_n^2 Y_0^2(r) | I, I \rangle \quad (2.14)$$

Because the operator who acts on the nuclear state carries 2 units of angular moment, this matrix element is zero when $I = 0, 1/2$. Because of the same reason the electric field

gradient $\frac{\partial^2 V}{\partial z^2}$ is zero when $J = 0, 1/2$ and depends on the electron configuration.

Because the electric field gradient $\frac{\partial^2 V}{\partial z^2}$ is isotope independent, the spectroscopic quadrupole moment Q_s can be measured if the B-factor and quadrupole moment of a reference isotope is known.

Total hyperfine splitting

The shift of the energy levels because of the quadrupole interaction is typically smaller than the shift because of the magnetic dipole interaction. The combined effect of the quadrupole interaction and the magnetic dipole interaction leads to an energy shift of the levels given by

$$\Delta E = -\frac{A}{2}C + B \frac{3C(C+1) - 4I(I+1)J(J+1)}{8(2I-1)I(2J-1)J}. \quad (2.15)$$

An illustration of the hyperfine splitting for a nucleus with spin $I = 3/2$ and $J = 2$ is given in figure 2.4.

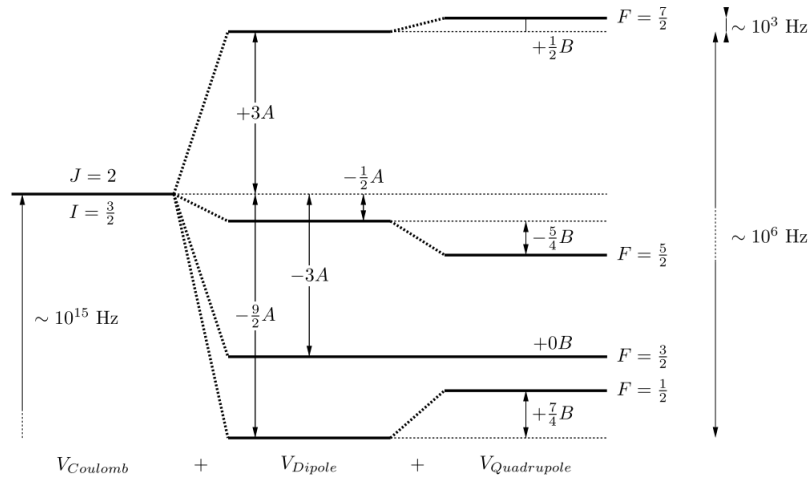


Figure 2.4: Hyperfine splitting for $I = 3/2$ and $J = 2$ of the energy level. From left to right the effect of the magnetic dipole interaction and the shift because of the quadrupole interaction is shown [29].

Chapter 3

Experimental set-up

The production of a radioactive ion beam has to deal with difficulties that stem from the very short half lives of the nuclei, the production of contaminants and the low production cross-section of the nuclei of interest. Therefore the production of a radioactive ion beam has to be fast, selective and efficient [30]. All these requirements led to the development of two complementary techniques for the production of a beam of exotic nuclei: the in-flight separation technique and the isotope separation on line (ISOL) technique [30]. The mercury isotopes are produced by the ISOL technique at the ISOLDE facility at CERN, the European Center for Nuclear Research. The ISOLDE facility is shown in figure 3.1

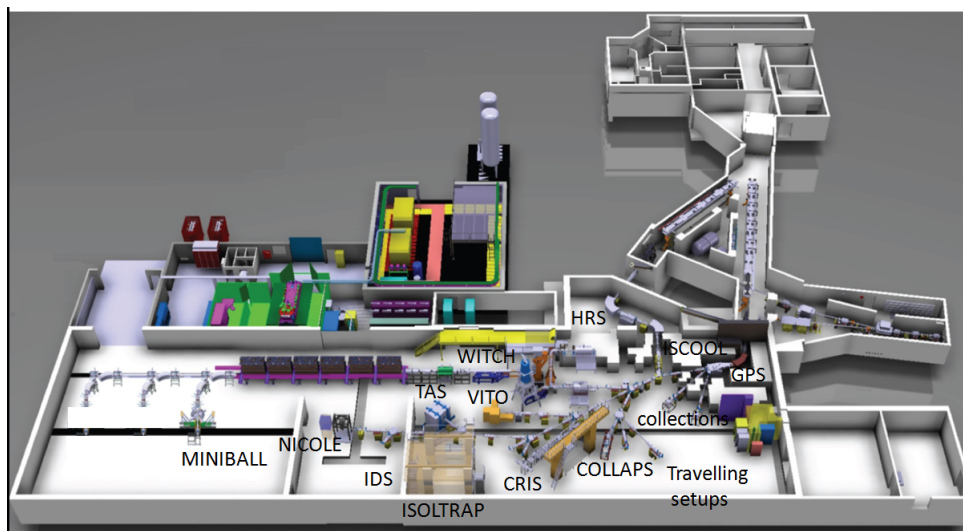


Figure 3.1: Layout of the ISOLDE facility [31].

The mercury isotopes are first produced in a target. After their production, they are stopped within the target itself, acting as a catcher. After stopping the reaction products diffuse out of the target before the isotopes of interest are ionized by lasers and mass separated by a dipole magnet. The resulting ion beam is sent to a detection system. This process is shown in figure 3.2. In what follows each of these steps will be discussed in more detail.

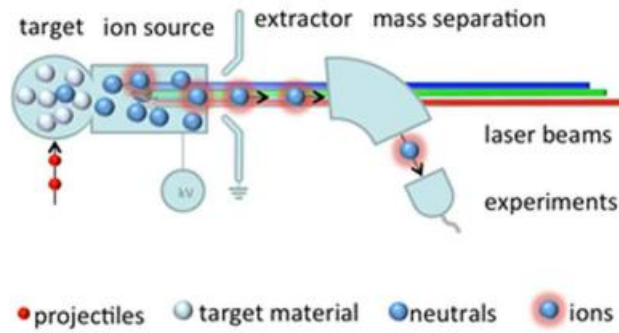


Figure 3.2: The mercury isotopes are first produced, thermalized, ionized, accelerated, mass separated and then detected in the detection system [32].

3.1 High temperature thick target and catcher

The mercury isotopes are produced by sending a high energy proton beam on to a molten lead target, which also serves as catcher for the reaction products. The protons at the ISOLDE facility were first accelerated by a linear accelerator up to an energy of 50 MeV and then they are sent to the Proton Synchrotron Booster (PSB). When the protons leave the PSB they have an energy of 1.4 GeV and can either be transported to ISOLDE, another CERN facility or further accelerated [33]. The proton beam consists of a super cycle of at most 42 different proton pulses. Each of these pulses have a length of $2.4 \mu\text{s}$ and are separated in time by 1.2 s.

When this high energy proton beam hits the target, three kinds of reactions can occur: fission, spallation and fragmentation. The neutron-deficient mercury isotopes are produced by a spallation reaction in which the lead nucleus loses two protons and several neutrons. In preparatory tests predating the experiment [15], it was investigated which target, molten lead or UC_x , is best suited for the experiment. In choosing between these materials one has to balance the highest production cross-section and the lowest amount of contaminants. It was concluded that molten lead and UC_x are equally good for the investigation of the neutron-deficient mercury isotopes down to ^{177}Hg . However, the proposal of the experiment did not only include the neutron-deficient mercury isotopes, but the neutron rich isotopes $^{207,208}\text{Hg}$ as well. Therefore the use of a molten lead target was preferred, because using the UC_x target leads to an overwhelming isobaric contamination of francium, which would have been produced via surface ionization.

After production and stopping, the isotopes are transported through the transfer line to the ion source. The transfer line and the ion source are also kept at a high temperature, in order to avoid losses of isotopes who remained sticking to the walls.

3.2 Ion source

Depending on the ionization potential of the element of interest, the required charge state and the selectivity three different ionization mechanisms can be used in ISOL systems: electron impact ionization, surface ionization and laser ionization. We are interested in the isotope shift of the mercury isotopes, hence laser ionization is the logical choice.

The process of laser ionization consists of a stepwise excitation of the isotopes by laser light. The isotopes are typically excited to the continuum, an auto-ionizing state or a Rydberg state. A Rydberg state is a highly excited state close to the continuum. In this case the final ionization happens by applying an electrical field, infra-red radiation or by atomic collisions. In most cases the ionization scheme consists of two or three steps. Most of these steps are resonant, which leads to a high ionization efficiency. Laser ionization is very selective but due to the high temperature of the cavity, isotopes of elements with a low ionization potential, like francium and thallium, are ionized as well. This can, after mass separation, lead to isobaric contamination [30].

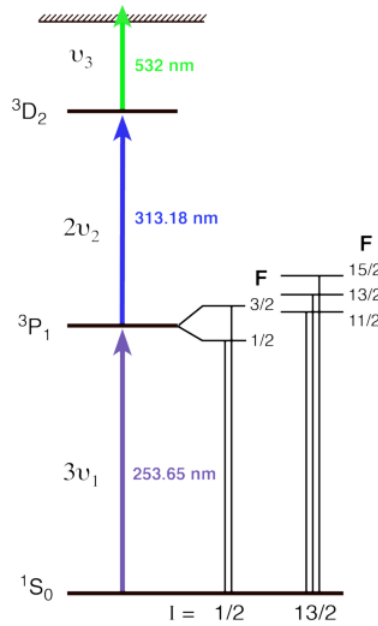


Figure 3.3: The laser ionization scheme for mercury and the hyperfine splitting. The hyperfine splitting is visible for the odd-mass isotopes. I is the nuclear spin for the ground state and isomeric state and F is the total spin [15].

The laser ionization scheme that is used to study the mercury isotopes is shown in figure 3.3. Three different lasers are used: a frequency tripled Ti:Sa laser is used for the first excitation step, a frequency doubled dye laser provides the light for the second step and the ionization to the continuum is completed by a Nd:YAG laser. During the experiment the frequency of the Ti:Sa laser is changed and the frequency of the two other lasers is kept constant. In this way we can study the isotope (isomer) shift of the $^1S_0 \rightarrow ^3P_1$ transition by using in source laser spectroscopy. Indeed, only when the laser light of the first laser is at resonance, the mercury isotopes are ionized and we detect their alpha decay at the Windmill detection system. In the end a plot of the intensity of the detected alpha particles versus the laser frequency is obtained. This plot reproduces the transitions shown in the lower part of figure 3.3. The resolution is limited by Doppler broadening because of the high temperature of the ion source. This width is for the mercury isotopes comparable with the laser bandwidth, which is ≈ 1.5 GHz after frequency tripling of the Ti:Sa laser light [15].

The ion source, called the Resonance Ionization Laser Ion Source (RILIS), is put on a positive high voltage of typical values between 40 and 60 kV [30]. The isotopes are then accelerated towards the mass separator.

3.3 Mass separation

The ion beam is mass separated by sending the beam through a dipole analyzing magnet. At ISOLDE two mass separators are available: the General Purpose Separator (GPS) and the High Resolution Separator (HRS). The GPS was used during the mercury experiment. Even when the mass resolution is very good, it is possible that after mass separation there are contaminants of neighboring masses present. The origin of this can be found in collisions with rest gas which induce a spread in energy of the ions. This change in energy leads to a change of direction. As a consequence there will be a relatively small amount of neighboring masses present in the ion beam after mass separation [33]. An example of a mass spectrum after mass analysis is shown in figure 3.4.

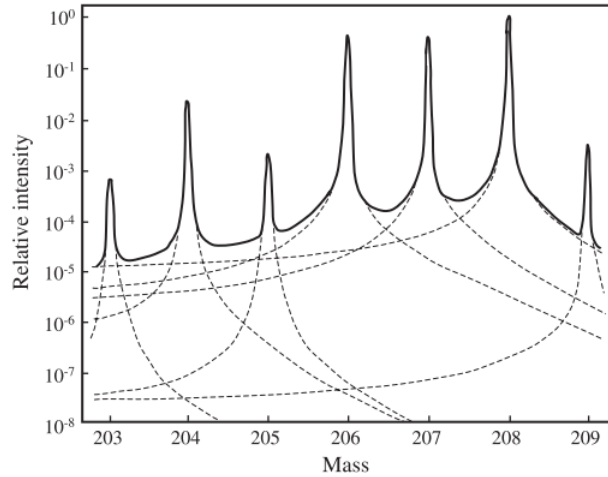


Figure 3.4: Spectrum of an ion beam obtained after mass separation [30].

3.4 Detection systems

In April 2015 experiments were conducted on 15 mercury isotopes ($^{177-185}\text{Hg}$, ^{198}Hg , ^{202}Hg , ^{203}Hg , $^{206-208}\text{Hg}$). A different ion counting technique was used depending on the mercury isotope, three different detection systems were available: a Faraday cup (FC), the Multi-reflection Time-Of-Flight (MR TOF) in combination with a multichannel-plate ion detector and the Windmill detection system (WM). A schematic representation of the detection system used for each mercury isotope is given in figure 3.5.

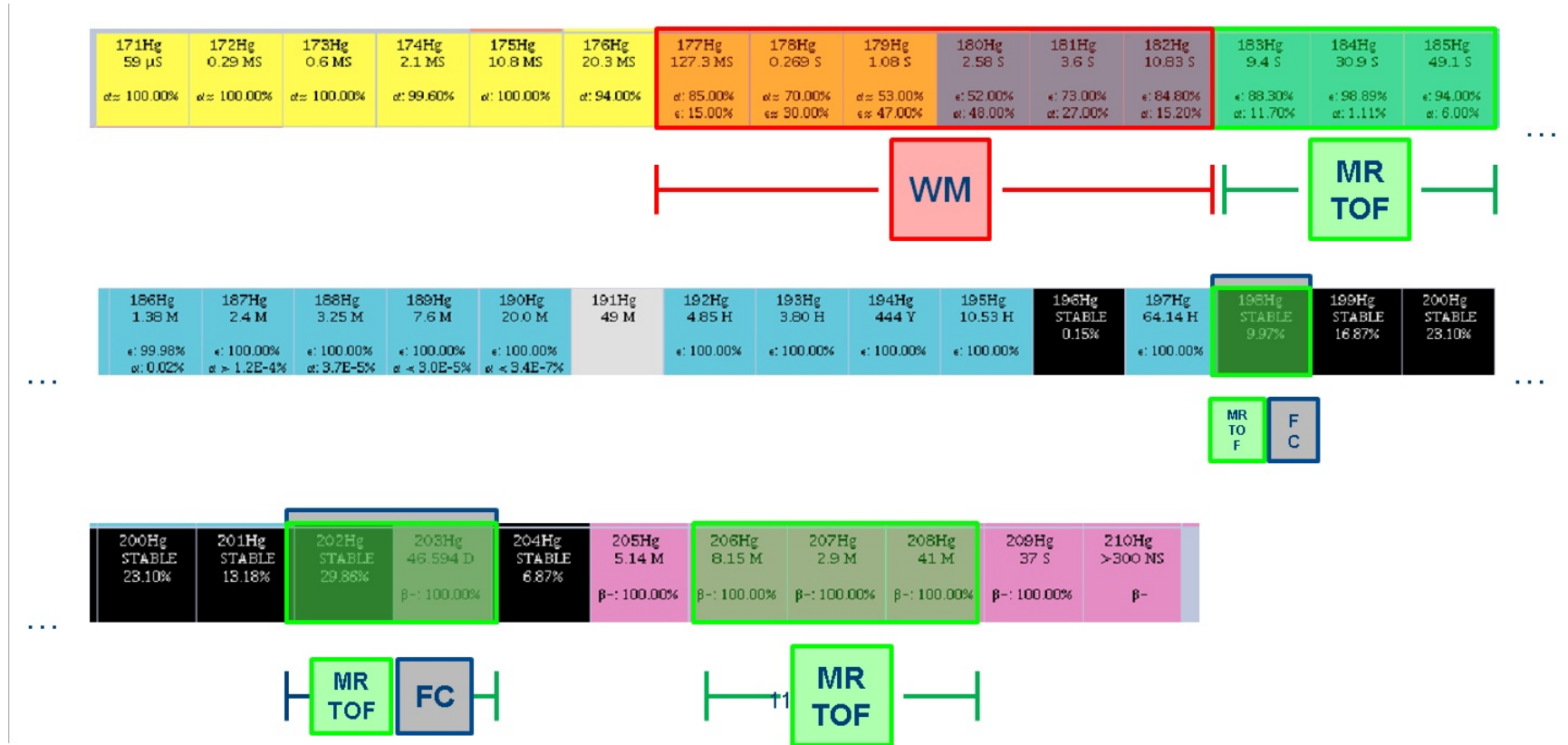


Figure 3.5: Representation of the detection system used for each mercury isotope. Three different detection systems were used: the Faraday cup (FC), the Multi-reflection Time-Of-Flight (MR TOF) and the Windmill detection system (WM).

3.4.1 Faraday cup

A Faraday cup is a metal cup which catches the incident ions. The contact with the metal surface neutralizes the ions and charges the cup. The resulting current can be measured and is proportional to the number of incident ions per second. This ion counting technique can only be used for isotopes with a high production yield. It relies on the GPS mass separator for reducing the background.

3.4.2 Multi-reflection Time-Of-Flight

The working principle of this set-up is explained briefly. In MR TOF ions undergo multiple reflections by electrostatic mirrors. Different ion bunches are mass separated through a difference in time-of-flight. The MR TOF mass analyzer at ISOLTRAP is shown in figure 3.6. It consists of two electrostatic mirrors of 160 mm long, which are separated by a 460 mm long in-trap lift electrode. The ions are injected by a radiofrequency quadrupole (RFQ) buncher. In a RFQ a negative potential gradient is present with the beginning set at a positive potential U . Bunching is achieved by periodically applying a potential barrier at the end of the RFQ which stops the ions for a particular time. Once enough ions are collected, the barrier is removed and the ions reach the MR TOF with a velocity $v_i \propto \sqrt{m_i/z_i}$ with m_i the mass and z_i the charge state. Thus, the time-of-flight $t_i \propto v_i^{-1} \propto \sqrt{m_i/z_i}$ inside the MR TOF will be mass and charge state dependent. Consequently, different masses can be resolved through their difference in time-of-flight. The ions undergo multiple reflections between the mirrors in order to increase the mass resolving power by extending the flight path. (The difference in time-of-flight increases linearly with the number of revolutions). The ions are injected and ejected with a total energy that exceeds the maximum potential energy inside the mirrors. Confinement is achieved by changing their total energy once they are in between the two mirrors. For this purpose an in-trap lift electrode is used. This electrode is at a positive potential U_{lift} when the ions are injected. The incoming ions will then have a kinetic energy $E_{\text{kin}} = ze(U - U_{\text{lift}})$. While they are inside the in-trap lift electrode, the potential of this electrode is lifted to zero. Therefore, the ions lose their potential energy and their total energy is now equal to E_{kin} which is lower than the energy needed to be able to leave the MR TOF. When the ions are ejected, the lift-electrode is set again to U_{lift} . After ejection the ions are counted by a multichannel-plate ion detector [34]. MR TOF was used in this experiment for the separation of stable and beta-decaying isotopes of mercury.

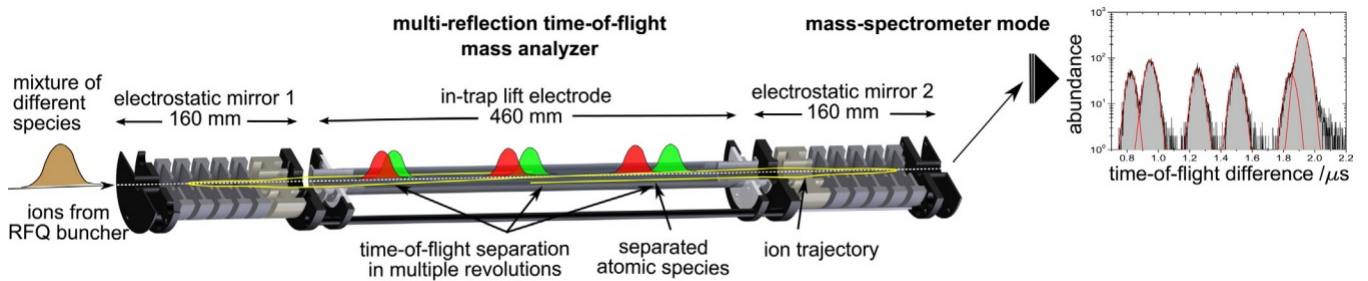


Figure 3.6: Sectional view of the MR TOF device at ISOLTRAP [34].

3.4.3 Windmill detection system

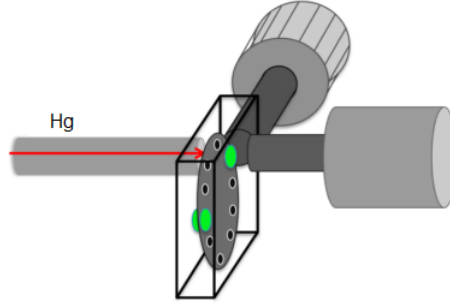


Figure 3.7: The Windmill detection system consists of a rotating wheel with carbon foils (black), Si detectors (green) and Ge detectors (the gray tubes outside the box) [33].

The Windmill detection system [33] is used to detect the characteristic alpha decay of mercury isotopes. Mercury isotopes are implanted in carbon foils. There are in total 10 thin carbon foils ($20 \mu\text{g}/\text{cm}^2$), which are located on a rotating wheel. The hyperfine spectrum can be constructed by gating on the correct alpha energy. The Windmill system is displayed in figures 3.7 and 3.8.

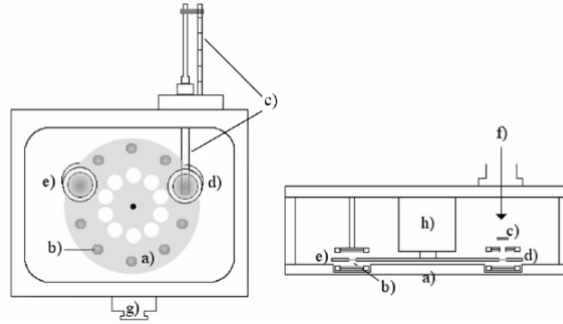


Figure 3.8: The front view (left) and top view (right) of the Windmill system displaying (a) rotating wheel, (b) carbon foil, (c) current measurement and collimator, (d) implantation station (with two surface barrier Si detectors), (e) decay station (with two PIPS Si detectors), (f) separator beam, (g) vacuum pump hole and (h) step motor [29].

Detectors

The Windmill system consists of four silicon detectors inside the Windmill vacuum chamber and two germanium detectors outside, as shown in figure 3.7. The names used to denote the detectors in this thesis are illustrated in figure 3.9.

Two Si detectors are placed at the implantation position; one annular detector through which the mercury ions pass before they are stopped in the carbon foils and one surface barrier detector. Outside the Windmill system there are two HPGe detectors directly behind the implantation point. The LEGe (Low Energy Germanium) detector, which is the one standing behind the vacuum chamber, is most efficient at energies around 60 keV. The Ge detector positioned at 90 degrees with respect to the ion beam direction has a

maximum photo-peak efficiency at roughly 200 keV. Surface barrier detectors are used at the implantation site because of their thinner backing. This allows X-ray and low-energy gamma rays to pass the Si detectors more easily.

On 144 degrees of the implantation station, measured with respect to the wheel center, the decay station is located. The decay station consists of two Passivated Implanted Planar Silicon (PIPS) detectors and is used to measure longer lived activity which is left on the carbon foil after beam implantation and rotation of the wheel.

Si and Ge detectors are semiconductor detectors. Most semiconductor detectors consist of a connection between a p-type and a n-type semiconductor. Because of the gradient in charge density, the electrons of the n-type material and the holes of the p-type material diffuse towards the other side of the junction. The electrons and holes recombine and this results in the creation of a region without a net charge, the depletion region. The depletion region can be enlarged by applying a reversed bias. Whenever a charged particle or photon enters the depletion region, electron-hole pairs are created. Those electrons and holes are accelerated by the electric field inside the depletion region. This results in a measurable voltage pulse, which height is proportional to the energy of the entering particle.

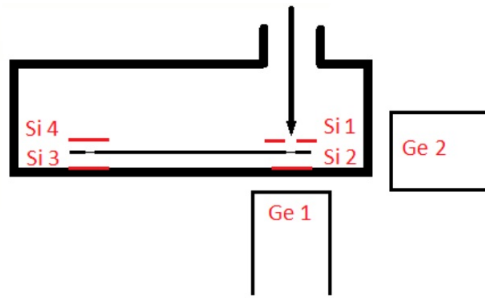


Figure 3.9: The naming for the detectors, as used in this thesis [33].

The detector specifications are given in the next two paragraphs.

Silicon detectors The silicon detectors are used to detect the alpha particles and the electrons resulting from the decay of radioactive isotopes implanted in the carbon foils. The detectors at the implementation station consist of an annular (Si1) and a full silicon (Si2) surface barrier detector, both made by Ortec. These detectors are placed at either side of the wheel.

Si1 has an active area of 450 mm^2 with a hole of 6 mm [35], through which the beam passes, included. The FWHM of Si1 was 35 keV for a typical alpha line during the run in April 2015. The absolute efficiency was 10%. This value is determined by using the source strength of a ^{241}Am calibration source.

Si2 has an active area of 300 mm^2 [35]. This detector had a FWHM of 33 keV for a typical alpha line during the run. The absolute efficiency was 15%. This value is determined by using the source strength of a ^{241}Am calibration source.

Two PIPS detectors, both made by Canberra, are placed at either side of the rotating wheel at the decay station. Both Si3 and Si4 have an active area of 300 mm^2 [36] and had a FWHM of 25 keV at an energy of 5480 keV, which is an alpha line of ^{181}Au . There

Detector	Model number	Serial number	Active area (mm ²)	Efficiency (%)	FWHM (keV)
Si1	TC-025-450-300-S	43 082H	450	10	35
Si2	TB-020-300-500	49 186B	300	15	33
Si3	PD 300-15-300 RM	74 256	300	-	25
Si4	PD 300-15-300 RM	74 257	300	-	25

Table 3.1: Properties of the used silicon detectors.

was not enough data available for these detectors during the mercury run, therefore an alpha line from ^{181}Au was used to determine the resolution. The isotopic chain of Au was measured after the mercury run. The efficiency could not be computed, due to a lack of efficiency measurements with ^{241}Am .

The properties of the silicon detectors during the mercury run are summarized in table 3.1.

In at least two articles [37] [38] it is stated that the distance between Si2 and the carbon foil is 3-4 mm. This results in an expected absolute efficiency around 30%, which is the solid angle at this distance. The efficiency of Si2 was missing a factor of 2 from what was expected. In November 2015 efficiency measurements on the Si detectors in the Windmill system were performed in order to solve this problem. The problem was solved by very accurately measuring the distance between Si2 and the Carbon foil. This distance was around 8 mm whereas it was previously thought to be 3-4 mm. The results of these measurements are discussed in Appendix C.

Germanium detectors Two High-Purity Germanium (HPGe) detectors, made by Canberra, are used in the experiment to detect X-rays and low-energy gamma rays. Ge1 which is placed parallel to the beam, is a Low Energy Germanium detector (LEGe) with an absolute efficiency of 0.3% at an energy of 1.1732 MeV. This efficiency is determined by using the source strength of a ^{60}Co calibration source. The FWHM was 1.2 keV at an energy of 85 keV during the run in April 2015.

Ge2, placed perpendicular to the beam, is a coaxial germanium detector with a relative efficiency of 75%. The relative efficiency is the efficiency of the germanium detector for the 1332 keV gamma line of a ^{60}Co 1.2×10^{-3} Bq source placed at a distance of 25 cm compared to the efficiency of a standard 3 inch x 3 inch NaI(Tl) detector [39]. The absolute efficiency of this detector was 0.5% at an energy of 1.1732 MeV. This value is determined by using the source strength of a ^{60}Co calibration source. The FWHM at an energy of 133 keV was 1.8 keV during this run.

The properties of the used germanium detectors are summarized in table 3.2.

Detector	Model number	Serial number	Photopeak efficiency (%) at 1.1732 MeV	FWHM (keV)
Ge1	771510	b 83547	0.3	1.2 at 85 keV
Ge2	GC7520	b 92022	0.5	1.8 at 133 keV

Table 3.2: Properties of the used germanium detectors.

Timing of data acquisition

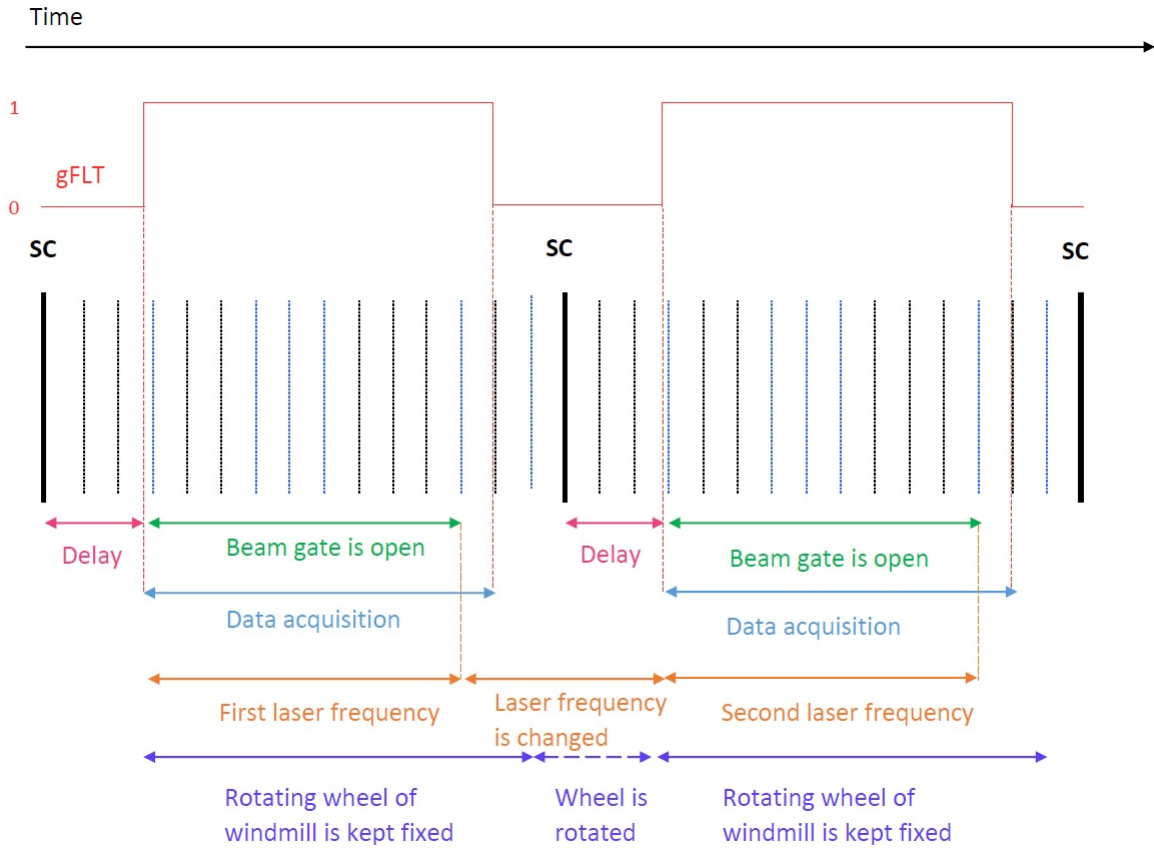


Figure 3.10: Schematic representation of the control logic of the set-up.

The rotation of the wheel is synchronized with the proton supercycle. Once the mercury ions are implanted in a carbon foil, the activity is measured for a certain period (depending on the production of the isotope of interest), before the foil is turned away and the remaining activity can be detected at the decay station while mercury ions can be implemented in an another carbon foil. During this rotation the laser frequency is changed.

A schematic representation of the control logic of the set-up is shown in figure 3.10.

A super cycle (SC) consists of multiple proton pulses. A dotted line in figure 3.10 is a representation for a proton pulse. Not every proton pulse within a super cycle was used for our experiment. The blue dotted lines are e.g. the ones which were used. The data acquisition for a new laser frequency is started after a fixed delay. In this way, there is enough time for the laser to stabilize at the new frequency and contamination from the previous frequency can be avoided. After the beam gate is closed, the data acquisition keeps going for some time. This is necessary in order to register the decay of the last implanted mercury ions. The laser frequency can be adjusted as soon as the beam gate is closed. Because rotating the wheel leads to some noise the wheel of the Windmill set-up is only rotated after the data acquisition has ended. Data acquisition is only started again when all veto signals entering an AND logic module are on a high level, signifying that all different parts of the experimental setup are ready such as the laser system, the data acquisition system (IGOR), etc... IGOR has a setting by which the data acquisition period can be controlled. This setting is called the gFLT. Whenever the signal of the gFLT

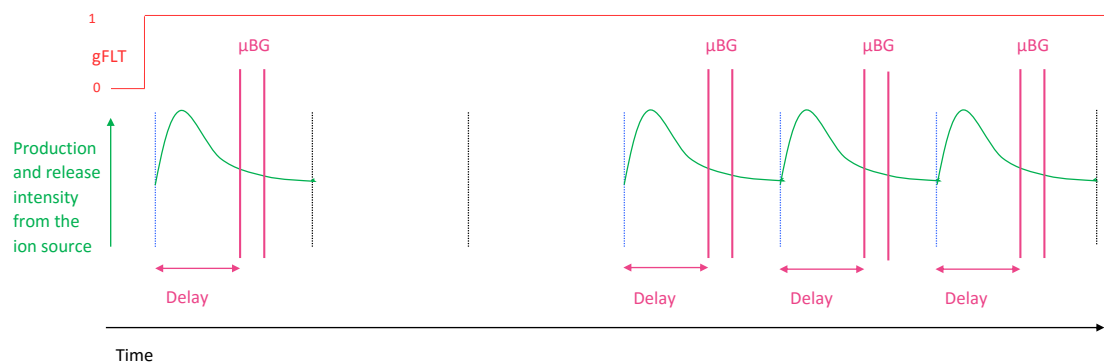


Figure 3.11: Working principle of a micro beam gate. Blue: proton pulses that are send to ISOLDE. Black: proton pulses that are not send to ISOLDE.

is equal to one, the data of the detectors which are connected to the gFLT, is saved. In some cases the production rate of the mercury ions was so high that the Ge detectors got saturated. This could be avoided by using a micro beam gate (μ BG). A μ BG is an electrostatic beam gate at the beginning of the beam line. Hereby, only the ions which leave the ion source after a fixed delay and during a certain period after the arrival of the proton pulses can enter the detection system. This principle is shown in figure 3.11.

Chapter 4

Hyperfine spectrum construction

In this thesis the way the hyperfine spectra (HFS) are constructed and analyzed for $^{181,182}\text{Hg}$ is discussed. The radioactive decay of these isotopes was measured with the Windmill detection system. Different scans were taken for each isotope to verify consistency. After checking the different files, the ones summarized in table 4.1 were used for further analysis.

Isotope	Name of the file
^{181}Hg	181Hg_scan_0271_to_0274
	181Hg_scan_0279_to_0280
	181Hg_scan_0362
^{182}Hg	182Hg_scan_0366
	182Hg_scan_0364

Table 4.1: The files which are analyzed for each isotope.

The isotope of interest is identified by its characteristic decay radiation. The HFS in which one plots a measure of the number of ions against the laser frequency can be made in two different ways: either by counting alpha particles from the radioactive decay or specific gamma rays. These spectra are extracted from the data in the following steps:

1. Selecting the alpha and X-ray/gamma energy conditions in the γ -ray spectra which will be used
2. Background subtraction for the HFS based on counting X-ray/gamma photons
3. Deadtime correction for both types of HFS

Each of these steps will be discussed in detail in the following sections. By following these steps, it is attempted to remove contamination of the spectra as much as possible as well as to correct for possible detector deadtime effects. Details e.g. extra spectra not included in this chapter are reported in Appendix A.

4.1 HFS based on counting alpha particles

Using this method of HFS creation, the HFS are extracted by counting the number of alpha particles belonging to the characteristic decay of the isotope of interest for each laser wavelength. This results in a high selectivity since different isotopes have other alpha energies and therefore the contribution from possible contamination can be removed.

4.1.1 Selecting the alpha lines

The first step consists of observing the energy spectra of the detectors at the implementation station. An example of such an energy spectra measured by Si2 for a scan of ^{181}Hg and ^{182}Hg is given in figure 4.1. The different alpha peaks observed in the spectrum have been identified and energy gates (indicated by the colored boxes in figure 4.1) were used to create the HFS displayed in figure 4.2. The number of alpha counts in a particular peak for Si1 and Si2 were summed at each laser wavelength and Poisson statistics was used to determine the errors on the number of counts.

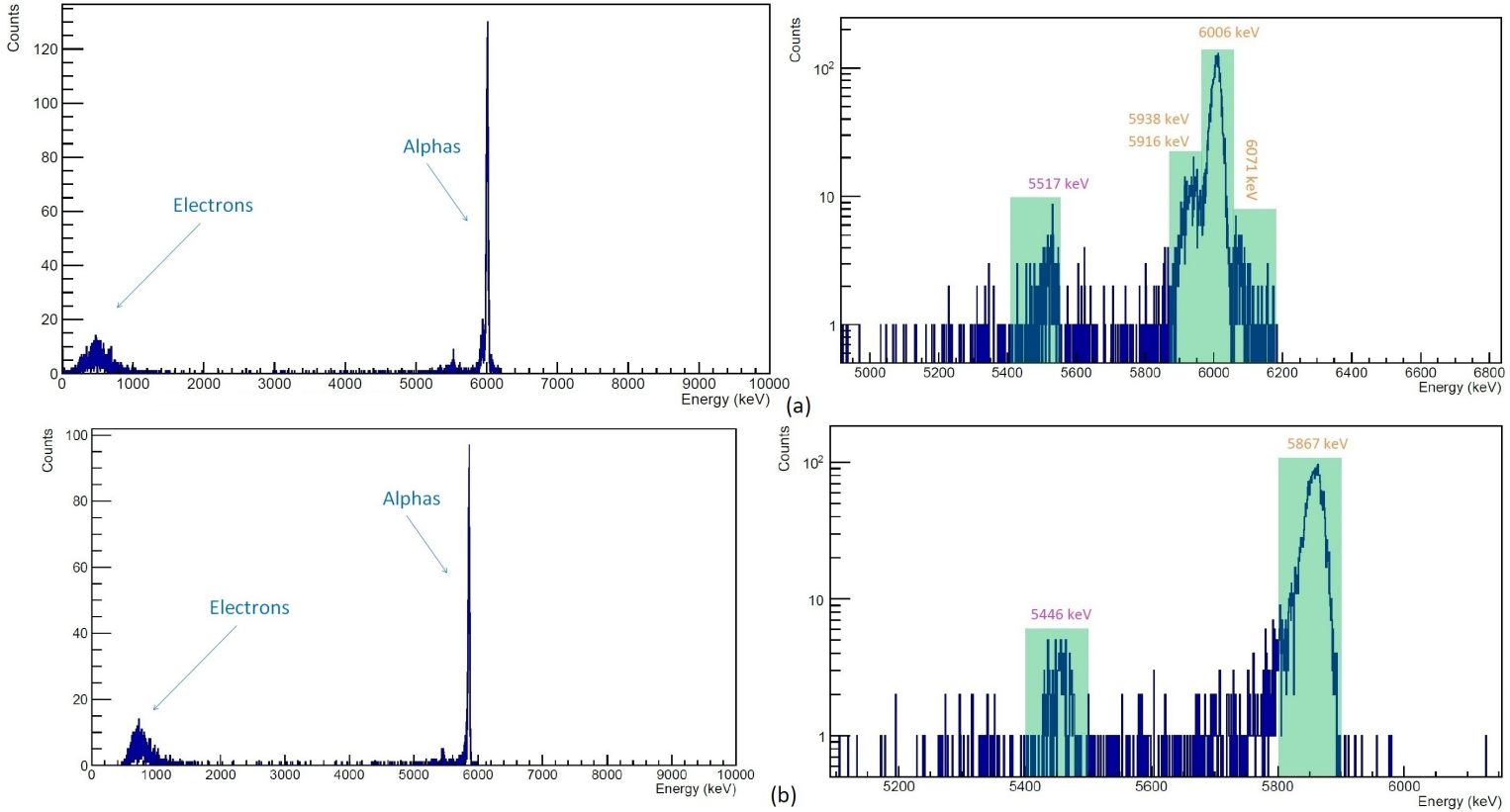


Figure 4.1: Energy spectra for Si2 of measurements (a) ^{181}Hg _scan_0279_to_0280 of ^{181}Hg and (b) ^{182}Hg _scan_0366 of ^{182}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay schemes of ^{181}Hg (figure 1.2a) and ^{182}Hg (figure 1.2b) in chapter 1.2.4. The green shaded areas represent the areas over which the integration is performed to make the HFS. The corresponding HFS are given in figure 4.2.

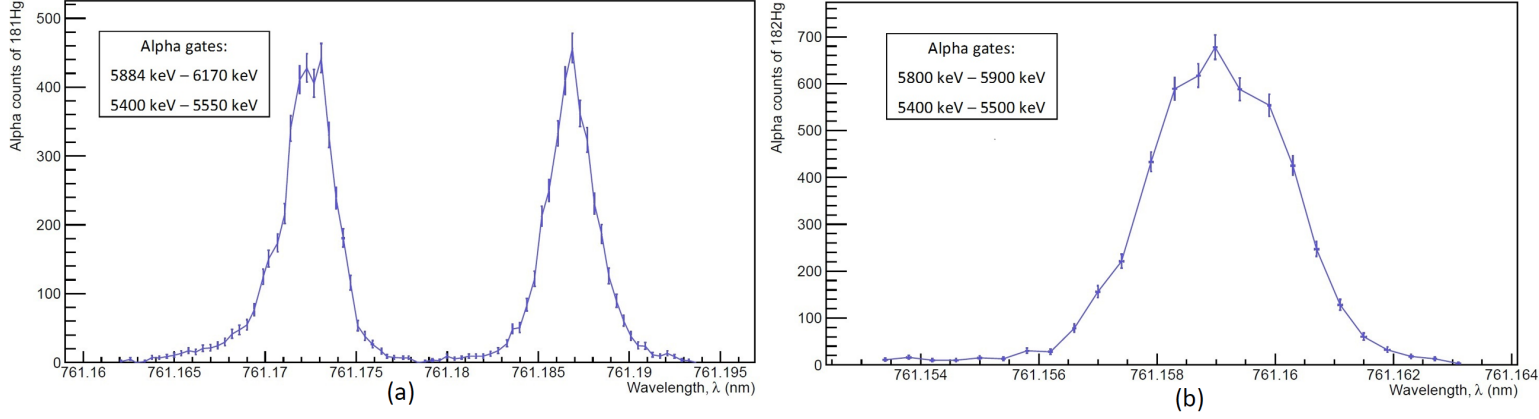


Figure 4.2: HFS based on counting alpha particles for measurements (a) 181Hg_scan_0279_to_0280 of ^{181}Hg and (b) 182Hg_scan_0366 of ^{182}Hg . The used alpha energy gates are written down in the upper box and are illustrated in figure 4.1.

4.2 HFS based on counting X- /gamma rays

These HFS are made by counting the number of X- /gamma rays belonging to the β -decay of $^{181,182}\text{Hg}$ for each laser wavelength. In contract to the alpha HFS (see figure 4.2), the HFS from gamma- or X-rays require background subtraction.

4.2.1 Selecting the X-ray/gamma line

Name of the file	LEGe gamma line (keV)	LEGe gamma energy gate (keV)	Ge2 gamma line (keV)	Ge2 gamma energy gate (keV)
181Hg_scan_0271_to_0274	42.5	41.2 - 44	none	none
181Hg_scan_0279_to_0280	42.5	40 - 44.6	none	none
181Hg_scan_0362	none	none	none	none
182Hg_scan_0366	129.5 217.6	129 - 130.4 215.9 - 218.5	217.6 413.5	216.6 - 218.9 412 - 414.5
182Hg_scan_0364	none	none	none	none

Table 4.2: Summary of which gamma rays and the corresponding energy gates are used for each measurement and each germanium detector.

The energy dependence of the gamma-ray detection efficiency is different for the LeGe (Ge1) and Ge2 (see chapter 3). Therefore different gamma gates to construct the HFS were used for the detectors (see table 4.2).

To identify the gamma transitions belonging to the decay of $^{181,182}\text{Hg}$, spectra taken when the lasers were set on-resonance are compared to spectra taken off-resonance. The on- and off-resonance range of the laser frequency was defined using the HFS spectra obtained using the alpha decay spectra. A correction for the different number of laser frequency steps (and thus the measurement time) was applied.

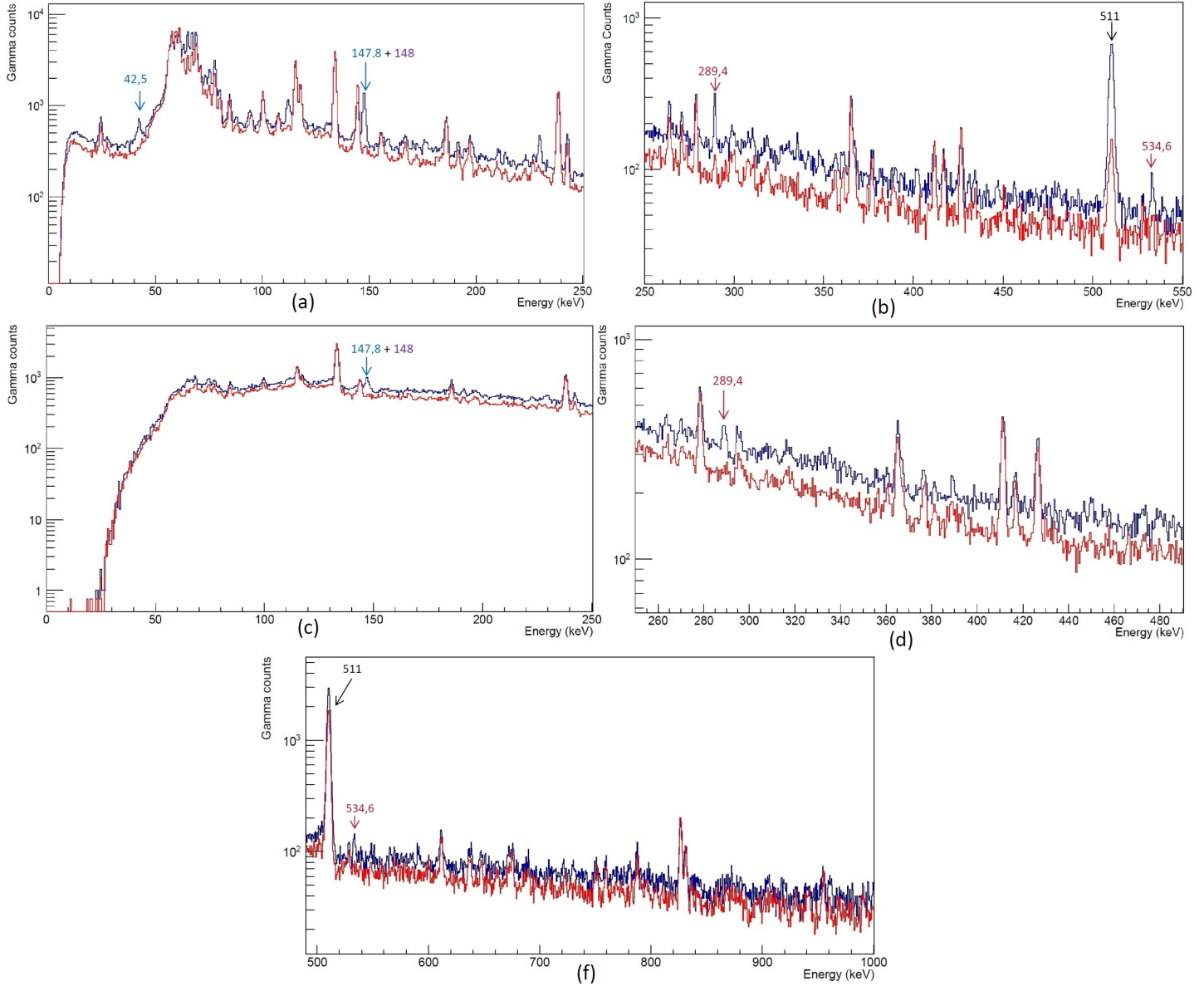


Figure 4.3: Energy spectrum on- (blue) and off- (red) resonance for measurement 181Hg_scan_0279_to_0280 of ^{181}Hg . (a)-(b) LEGe (c)-(e) Ge2. The gamma rays belonging to the β -decay chain of ^{181}Hg are identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{181}Hg (figure 1.2a).

Gamma rays that were enhanced in the laser on-resonance spectra are identified as belonging to the β -decay chain of $^{181,182}\text{Hg}$ - $^{181,182}\text{Au}$ - $^{181,182}\text{Pt}$ or the β -decay of $^{177,178}\text{Pt}$ which is the alpha decaying daughter of $^{181,182}\text{Hg}$. Figures 4.3 and 4.4 show a comparison of the gamma ray spectra for lasers on- and off-resonance for a measurement of ^{181}Hg and ^{182}Hg , respectively. The enhanced transitions in the laser on-resonance spectrum identified as belonging to the $^{181,182}\text{Hg}$ decay chain (see decay scheme of ^{181}Hg in figure 1.2a and of ^{182}Hg in figure 1.2b) are indicated. To construct the HFS spectra, however, the gamma and X-ray transition of the decay from the daughter nuclei can not be used for further analysis.

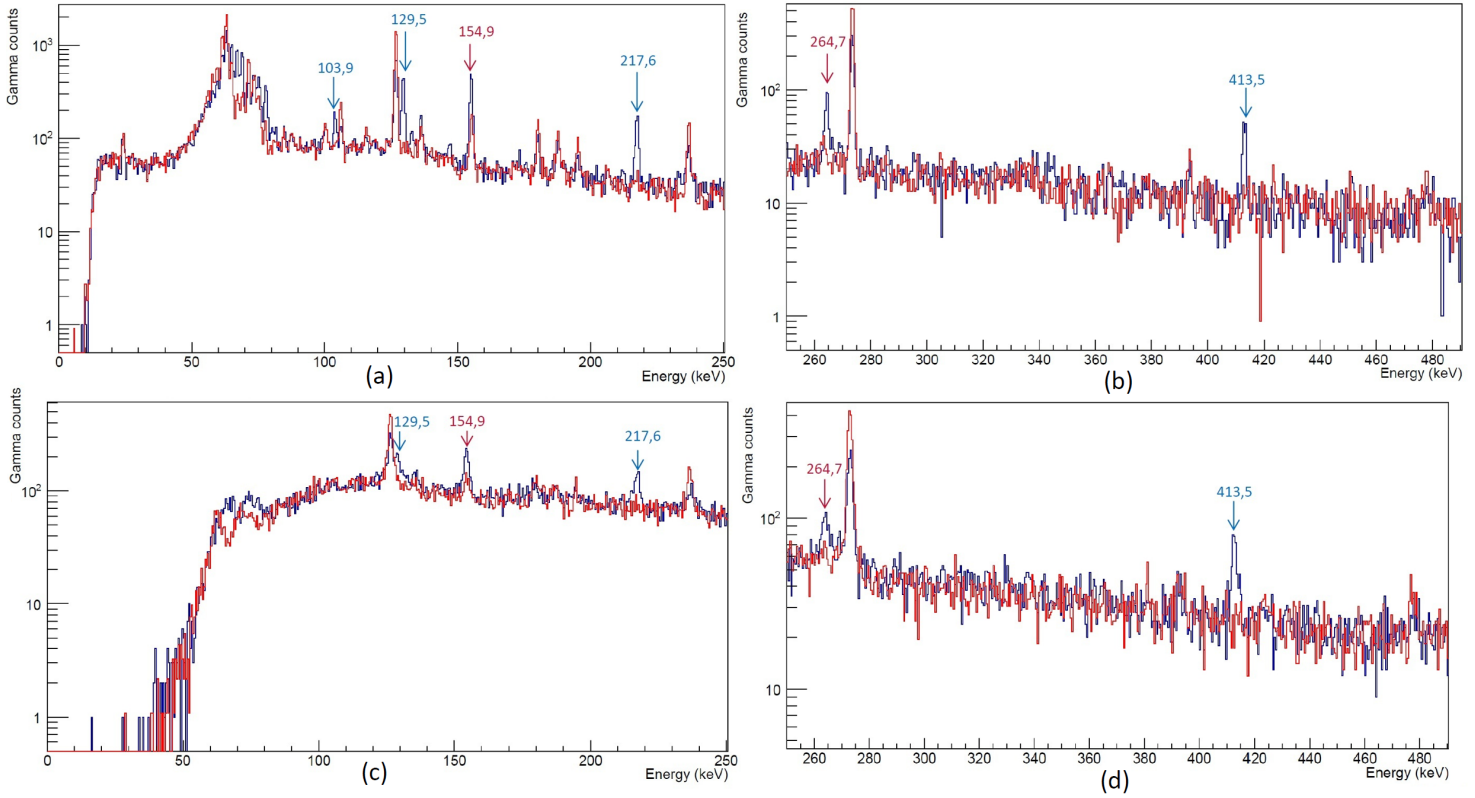


Figure 4.4: Energy spectrum on- (blue) and off- (red) resonance for measurement 182Hg_scan_0366 of ^{182}Hg . (a)-(b) LEGe (c)-(d) Ge2. The gamma rays belonging to the β -decay chain of ^{182}Hg are identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{182}Hg (figure 1.2b).

A small part of the radioactive beam is collected at the diaphragm in front of the carbon foil of the Windmill system. This results in a source that is not removed when the wheel turns to place a fresh carbon foil at the implantation position. Alpha radiation emitted from this source is not detected by the alpha particle detectors, but gamma and X-rays are detected by the germanium detectors. For example, the 154.9 keV transition of the β -decay of ^{182}Au is present in the off-resonance spectrum (see figure 4.4). Therefore only gamma rays from the β -decay of $^{181,182}\text{Hg}$ are suitable to make HFS.

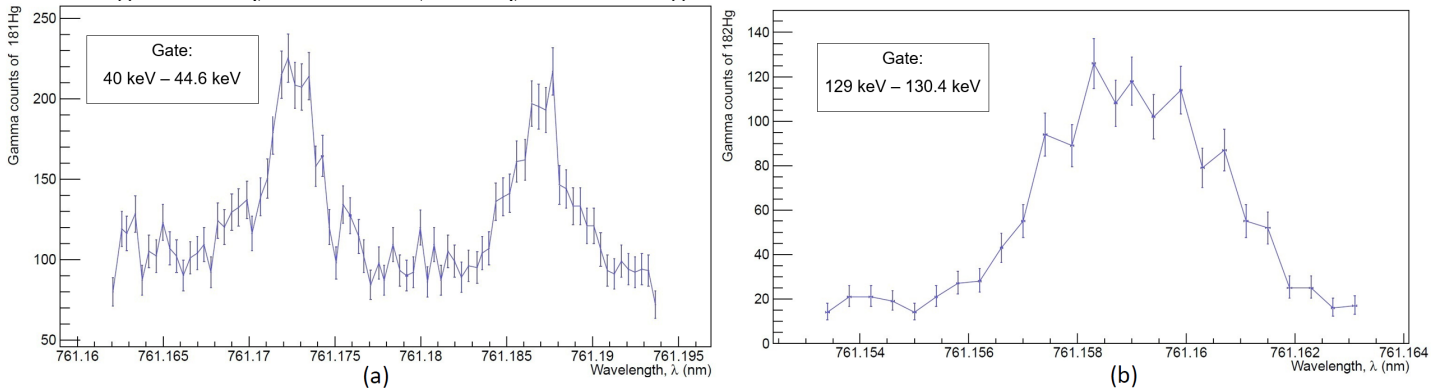


Figure 4.5: Examples of gamma HFS taken by LEGe for measurements (a) 181Hg_scan_0279_to_0280 of ^{181}Hg obtained by gating on the 42.5 keV line and (b) 182Hg_scan_0366 of ^{182}Hg obtained by gating on the 129.5 keV line. The used energy gates are displayed in the upper box.

The gamma rays used to construct the HFS for $^{181,182}\text{Hg}$ are given in table 4.2. An example of such a HFS for a measurement of ^{181}Hg and ^{182}Hg is given in figure 4.5.

4.2.2 Background subtraction

Gamma lines observed in the spectra are on top of a background due to Compton scattering of the gamma rays in the germanium crystal. This translates into a lower peak-to-background ratio in the gamma HFS compared to the alpha HFS (compare figure 4.5 with figure 4.2). Therefore a background subtraction procedure (illustrated in figure 4.6) was applied.

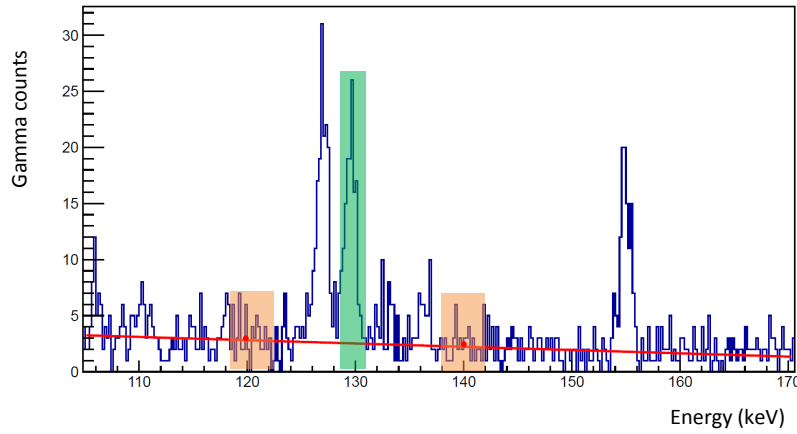


Figure 4.6: Illustration of the used technique for background subtraction. Green: the area over which is integrated to make the HFS obtained by gating on the 129.5 keV line taken with LEGe in measurement 182Hg_scan_0366 of ^{182}Hg . Orange: the chosen background areas left and right from the peak. Red: simulated background level for the fifteenth laser step.

In the analysis software program a background region has to be defined left and right of each peak. For every laser step the chosen background windows are reduced to two background points; one left and one right from the peak. The energy position (= x-position in the energy spectrum) of these points is determined by the weighted average of the bins in those windows. The weight given to each bin is the content of the bin. The gamma counts of these points (= y-position in the energy spectra) are the weighted averages of the counts (with Gaussian errors) in each bin in those windows. (The simplification of Gaussian instead of Poisson statistics is sufficient on the desired level of background correction). The error on the y-position is the error that results from this weighted mean calculation. There is no error given to the x-position. The line fitted between those two points is used to reproduce the background level in the γ -ray spectrum in each laser step. The error in the fit parameters is translated into an uncertainty in the background level which is included in the displayed error bars in the HFS with background subtraction. A comparison of the HFS obtained by gating on the 129.5 keV line from the β -decay of ^{182}Hg before and after background subtraction is displayed in figure 4.7. This background subtraction procedure had a minor influence on the quality of the HFS.

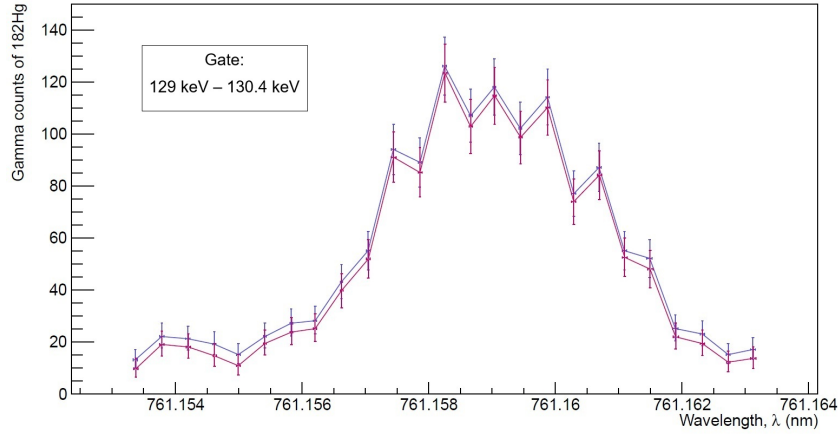


Figure 4.7: HFS obtained by gating on the 129.5 keV line from the β -decay of ^{182}Hg taken by LEGe of measurement 182Hg_scan_0366, before (blue) and after (pink) background subtraction.

4.3 Deadtime correction

The detector signals from the Windmill system are acquired digitally, using DGF modules from XIA. Each DGF module can store about 1000 events. When one of the modules has a full buffer (or at the end of a measurement cycle) a readout is performed which takes about 80 ms to complete. When the buffers of the DGF modules are emptied, the acquisition of incoming signals is blocked [40]. This results in deadtime of the acquisition system. Because the event rate within the Ge detectors is larger than in the Si detectors, the data acquisition buffer is filled up faster by the LEGe and Ge2 than by the other detectors.

In the experiment, a 100 Hz pulser was used to correct for acquisition deadtime by comparing the amount of pulser pulses in every laser step with those of the first laser step. By dividing the amount of detected alpha particles/gamma rays in each laser step by the ratio of the number of pulser pulses observed in the present laser step to the number of pulser pulses observed in the first laser step, the detected counts in every laser step are corrected with respect to the same reference (the first laser frequency).

For measurements 181Hg_scan.0271_to.0274 and 181Hg_scan.0279_to.0290 of ^{181}Hg it had to be taken into account that the pulser was not connected to the gFLT signal (explained in section 3.4.3). Consequently in some laser steps a double amount of pulser pulses was observed than expected from the data acquisition time. This is explained by the fact that sometimes a SC was missed when a veto-signal indicated that one of the components of the experimental setup was not yet ready (see figure 3.10). Missing a SC means that no new data acquisition period (indicated with the gFLT signal being equal to one) has started with the arrival of a new SC e.g. the second one in figure 3.10, but only when the next SC arrived. While the data acquisition for the detectors had already stopped with the drop of the gFLT signal at the end of the first SC, the pulser pulses kept being registered for the laser frequency belonging to the first SC until the arrival of the third SC. A small adjustment taking into account that not the total amount of pulser pulses have to be compared, but only the number of pulser pulses that arrived from the beginning of the laser step till the end of the data acquisition time, was made to the

program in order to perform deadtime correction for those two measurements.

The conclusion of the deadtime analysis is that only one of the analyzed scans suffered from acquisition deadtime, namely measurement 181Hg_scan_0271_to_0274 of ^{181}Hg . The deadtime corrected HFS are shown in figure 4.8. The laser frequency was scanned from right to left (starting at 761.195 nm). There is more acquisition deadtime on-resonance than off-resonance since the buffers of the DGF modules are filled in a shorter time period on-resonance. The deadtime correction is larger left from the peak than right, since the germanium detectors are still collecting all the information of the subsequent decay of ^{181}Hg , which was substantially produced at resonance.

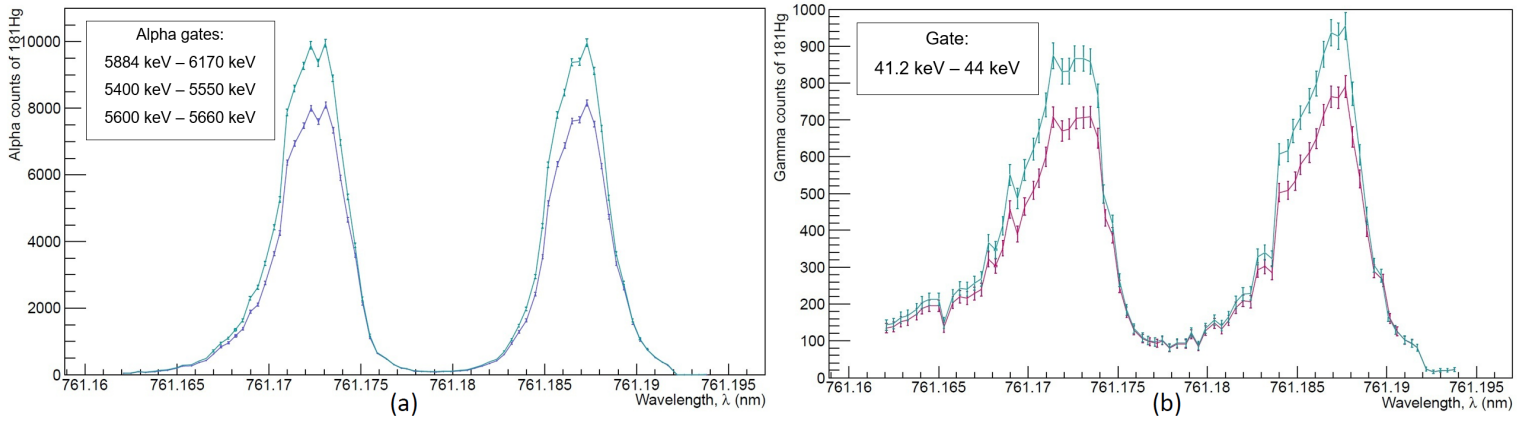


Figure 4.8: HFS with and without deadtime correction for measurement 181Hg_scan_0271_to_0274 of ^{181}Hg . The first five points from the left have to be removed since the Faraday cup was still in during these measurements. The used energy gates are displayed in the upper boxes. (a) Alpha HFS. Blue: without deadtime correction. Green: after deadtime correction. (b) Gamma HFS obtained by gating on the 42.5 keV line of the β -decay of ^{181}Hg taken by LEGe. This HFS has a big tail in both resonance peaks. Since these tails appear after the resonance frequencies, it is believed that they originate from remaining activity on the collimator. Pink: with background subtraction. Green: after background subtraction and deadtime correction.

A verification of the used deadtime correction in this work was performed by comparing the results with those obtained with the program of the collaboration in York (see figure 4.9). It can be concluded that the results agree very well. This gives confidence in the performed deadtime correction in this work.

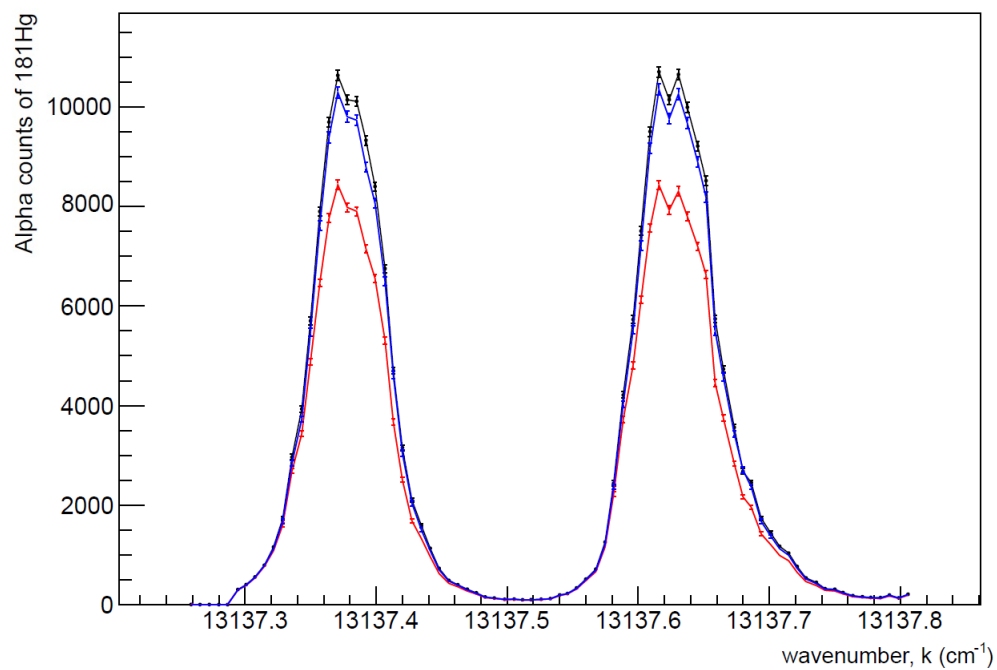


Figure 4.9: Comparison between deadtime correction results. Shown is the HFS based on counting alpha particles for measurement 181Hg_scan_0271_to_0274 of ^{181}Hg . Red: without deadtime correction. Blue: after deadtime correction with the program used in this work. Black: after deadtime correction using the program of the collaboration.

Chapter 5

Analysis of the hyperfine structure spectra

In this chapter the results of the HFS fit analysis are presented. The center of gravity is determined for $^{181,182}\text{Hg}$ and ^{198}Hg , the latter is the reference isotope used. The isotope shift $\delta\nu^{198\ 181,182}$ is evaluated and compared with the literature values in order to verify the reliability of extracting the differences in mean square charge radii using the in-source laser spectroscopy method. The difference in mean square charge radius $\delta \langle r^2 \rangle^{198\ 181,182}$ is calculated with the aid of the isotope shift $\delta\nu^{198\ 181,182}$ and from this value the nuclear model dependent difference in mean square deformation parameter $\delta \langle \beta^2 \rangle^{198\ 181,182}$ is evaluated.

5.1 Determination of the center of gravity

5.1.1 Fit algorithms

The HFS are fitted with the use of the SATLAS analysis program [41]. SATLAS stands for Statistical Analysis Toolbox for Laser Spectroscopy and is especially designed for the analysis of data gathered from laser spectroscopy experiments. Both Chi-Square and Maximum Likelihood Estimation fitting routines are available.

Chi-Square (χ^2) fitting is based on the idea that the best fit is the one in which the distance between the data points and the fit function is minimal. This distance can be optimized by minimizing the cost function

$$\chi^2 = \sum_i \left(\frac{y_i - f_{HFS}(x_i)}{\sigma_i} \right)^2.$$

The sum is taken over every data point i , y_i and x_i are the corresponding coordinates, σ_i is the error on y_i and f_{HFS} is the fit function. If it is also necessary to include an error on the x-values, this cost function has to be modified. However for all the measurements the statistical spread of the laser frequency measurement was around 10 MHz, while the spacing between successive laser steps was about 200 MHz. Therefore it was not necessary to include the x-error.

In order to determine the error on a fit parameter, the value of χ^2 is calculated for different fixed values of this parameter around the optimum value while refitting the rest of the

parameters. In the ideal case the resulting function will be a parabola with the minimum at the optimum value. The error on the fit parameter is determined by the interval in which the minimal χ^2 value has increased by one. These errors are then scaled in order to account for the goodness of the fit. The ideal fit has a reduced Chi-Square χ_{red}^2 , which is the χ^2 -value divided by the number of degrees of freedom, equal to one and fitting residuals randomly scattered around zero. These scaled errors are estimated from the covariance matrix. An alternative way would be to refit the HFS with the y-errors multiplied by the square root of the χ^2 -value. In this way the χ_{red}^2 is forced to be one and the errors determined by using the parabolas should give the same value as those estimated from the covariance matrix. This was checked for one HFS and the two methods gave indeed the same result.

The maximum likelihood estimation (mle) is a method to seek the probability distribution that makes the observed data most likely [42], which means that the best fit function is the one that maximizes the likelihood function $\mathcal{L}(\vec{\theta}|x, y, \sigma)$. $\mathcal{L}(\vec{\theta}|x, y, \sigma)$ is the probability that the data set (x, y, σ) is generated by the probability distribution characterized by the set of parameters $(\vec{\theta})$. SATLAS minimizes $-\log\mathcal{L}(\vec{\theta}|x, y, \sigma)$ while taking Poisson errors on the y-values. The error on a fit parameter is determined by calculating $-\log\mathcal{L}(\vec{\theta}|x, y, \sigma)$ for different fixed values of this parameter around the optimum value while refitting the rest of the parameters. In the ideal case the resulting function will be a parabola with the minimum at the optimum value. The error on the fit parameter is determined by the interval in which $-\log\mathcal{L}(\vec{\theta}|x, y, \sigma)$ has increased by 1/2. Mle is preferred over Chi-Square for fits with low statistics.

Every HFS of the Hg data is fitted with both the Chi-Square and the mle method to check consistency. The results of the Chi-Square method were used as initial conditions for the mle fit, since mle is much more sensitive to the initial conditions. Both methods are consistent, as discussed in sections 5.1.2 and 5.1.3. The statistics is high enough to rely on the results of the χ^2 -fitting for the further analysis. The difference in error bars for the two methods can be explained by the fact that the χ^2 -fitting takes into account the goodness of the fit, contrary to mle.

The fit parameters are a constant background level, the centroid position (which is the COG), the amplitudes, the full width at half maxima of the Lorentzian (FWHML) and Gaussian (FWHMG) profile and the hyperfine parameters.

5.1.2 Fit results for ^{182}Hg

The spin I of the even-even ^{182}Hg nucleus is equal to zero. Consequently the magnetic dipole moment μ and spectroscopic quadrupole moment Q_s are also zero. Therefore, as discussed in section 2.2.2, the magnetic dipole and quadrupole interaction parameters (A and B) of the upper and lower level of the transition are fixed to zero in the fitting procedures. This results in one resonance line in the HFS spectra.

Two measurements are available: 182Hg_scan.0364 and 182Hg_scan.0366, referred to as scan 0364 and 0366, of which the laser frequency was scanned in opposite directions.

Measurement number	0364	0366
Profile	Voigt	Gaussian
χ^2_{red}	76.6	2.3
Center of gravity (MHz)	1181589385 (66)	1181589291 (42)

Table 5.1: Summary of fit results obtained for χ^2 -fitting of the HFS based on counting alpha particles for the two measurements of ^{182}Hg . The displayed errors are corrected for the χ^2_{red} -value. These values for the center of gravity are used for the further analysis. The error intervals of the results for the center of gravity of the two measurements overlap and the results agree within two sigma.

HFS based on counting alpha particles

Measurement 0364 The alpha HFS for measurement 0364 was fitted with a Voigt profile. The corresponding correlation plot obtained by performing a χ^2 -fit is shown in figure 5.1. The plots on the diagonal are the calculated χ^2 -values for different values of one parameter while keeping all the others constant. From these plots it is clear that a minimum for the χ^2 -value in parameter space could be found and that the error bars on the fit parameters are reliable. The given error bars are the ones without correcting for the goodness of the fit. On the off-diagonal contour plots with 1,2 and 3 sigma contours are displayed. The χ^2 -fit is given in figure 5.2.

The fit results for the center of gravity have to be multiplied with three since the laser frequency is tripled after the wavelength measurement (see figure 3.3). The values for the center of gravity, after tripling, are shown in figure 5.3. Both fitting procedures are in good agreement. Since the error bars that are shown on the correlation plot are close to those obtained from the mle-fitting, the difference in error bars between the mle-result and the χ^2 -result can be explained by the reduced Chi-Square value ($\chi^2_{red} = 76.6$).

Measurement 0366 The HFS based on counting alpha particles for measurement 0366 was first fitted with a Voigt profile. However from the correlation plot of the χ^2 -fit (see figure B.2 in Appendix B) it could be concluded that the Voigt profile is not the suited one to use. Therefore a Gaussian profile is used. The corresponding correlation plot was reliable. The χ^2 -fit is displayed in figure 5.2.

The results for the center of gravity, after tripling, are shown in figure 5.3. Both fitting procedures are in good agreement. The smaller difference in error bars (compared to measurement 0364) can be explained by the reduced Chi-Square value ($\chi^2_{red} = 2.3$), which is closer to one.

The Chi-Square fitting results will be used in the further analysis and are summarized in table 5.1. The results for the center of gravity of the two measurements of ^{182}Hg agree within two sigma, as indicated in figure 5.3.

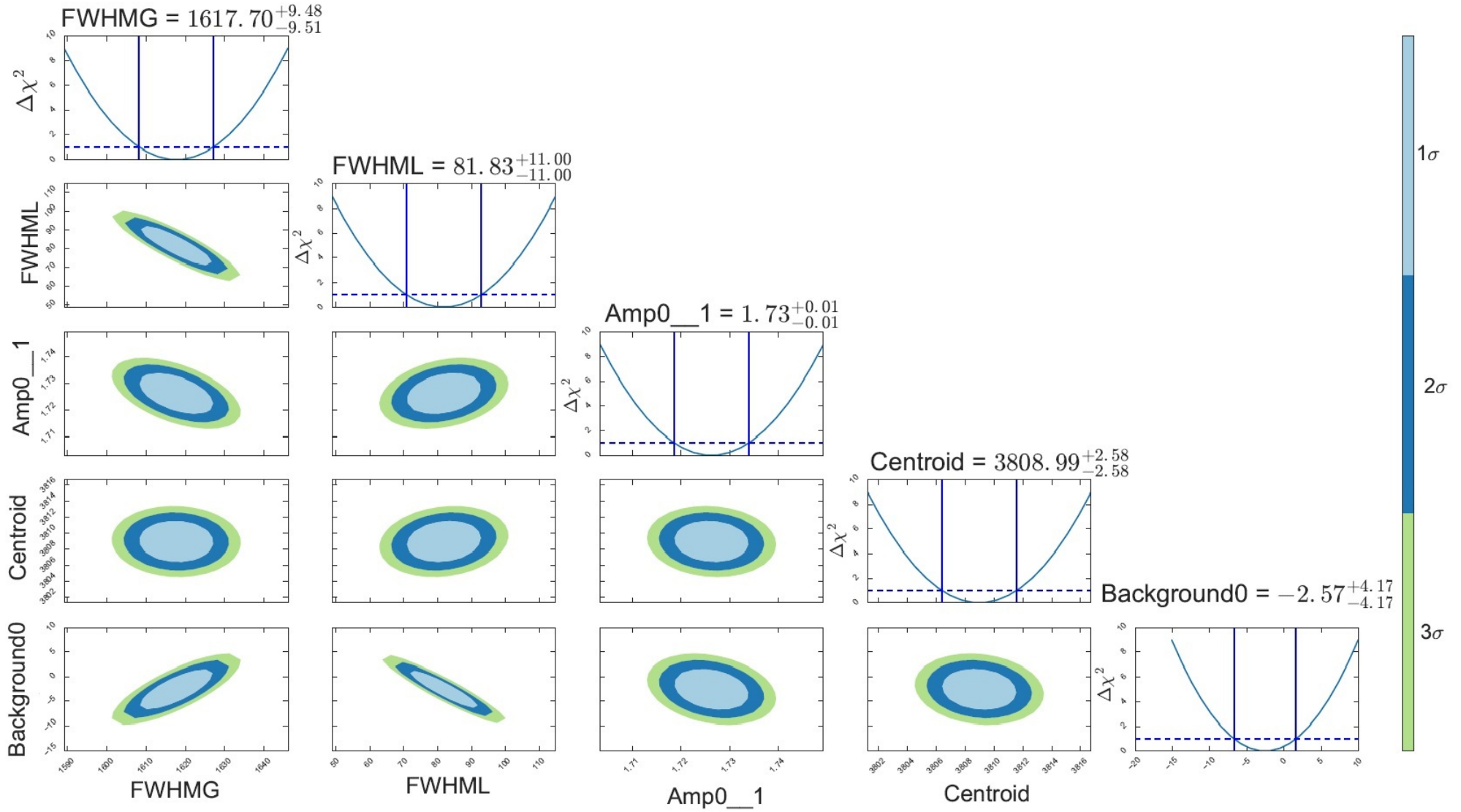


Figure 5.1: Correlation plot for measurement 0364 obtained by performing a χ^2 -fit with a Voigt profile. The plots on the diagonal are the calculated χ^2 -values for different fixed values of one parameter while refitting the rest of the parameters. In the ideal case, these plots should be parabolas. From these plots it is clear that a minimum for the χ^2 -value in parameter space could be found and that the error bars on the fit parameters are reliable. The shown error bars are the ones without correcting for the goodness of the fit. On the off-diagonal contour plots with 1,2 and 3 sigma contours are displayed.

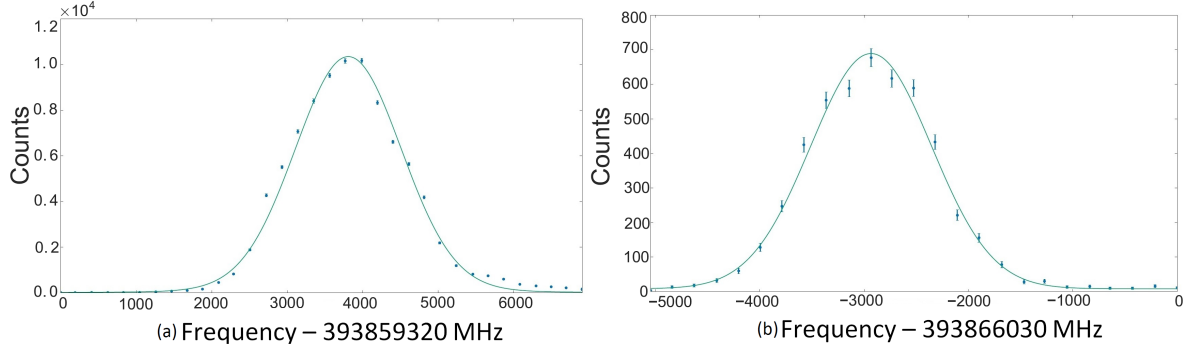


Figure 5.2: Fitted alpha HFS form the two measurements of ^{182}Hg . (a) Measurement 0364: the HFS is fitted with a Voigt profile ($\chi^2_{red} = 76.6$). (b) Measurement 0366: the HFS is fitted with a Gaussian profile ($\chi^2_{red} = 2.3$). The frequency difference with the first measured frequency is displayed. The results are summarized in table 5.1.

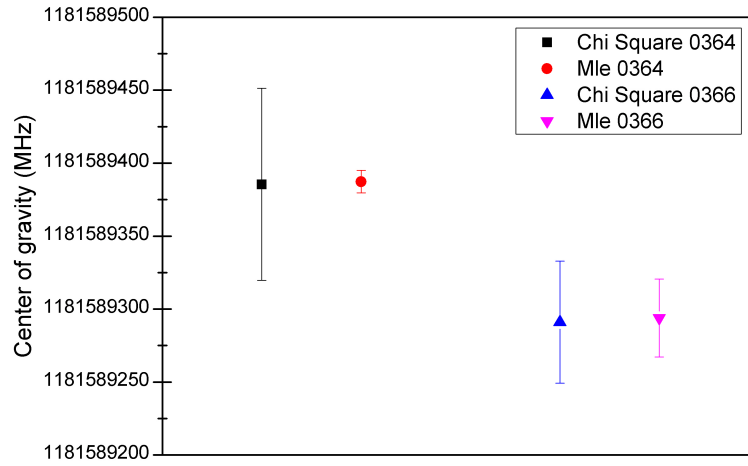


Figure 5.3: Results for the center of gravity of ^{182}Hg , after tripling the fit values, for both χ^2 -fitting (including χ^2_{red} -correction) and mle-fitting of the alpha HFS form the two measurements (0364 and 0366).

HFS based on counting X-/ gamma rays

Measurement 0364 For this measurement there are no gamma HFS available (see Appendix A.2.2).

Measurement 0366 There are 4 HFS based on counting photons available: two measured by the LEGe detector and two by Ge2. Based on excessive computational time for the correlation plot calculation and unreliable error results for a χ^2 -fit using a Voigt profile, a Gaussian profile is used instead. The fitted HFS are reported in appendix B in figure B.1.

The results for the center of gravity are displayed together with the those obtained from the alpha HFS in figure 5.1.2. The result for the 129.5 keV energy gamma measured by the LEGe detector is in agreement with the center of gravity determined by the alpha HFS. The results of the other gamma HFS match less. The larger error bars on the fit

values for gamma HFS result from the much lower statistics in these spectra. The lower statistics can be explained by the fact that these gamma lines have a lower intensity. It can be concluded that there is an agreement between the results for the alpha and gamma HFS except for the last three gamma HFS. The result of the alpha HFS agrees within two sigma with the values of these three gamma HFS. A possible contamination in the gamma ray spectra or higher background conditions for the germanium detector might be considered. A final conclusion can not be made at this point. In the following the data obtained from the HFS created from the alpha decay spectra will be used (see table 5.1).

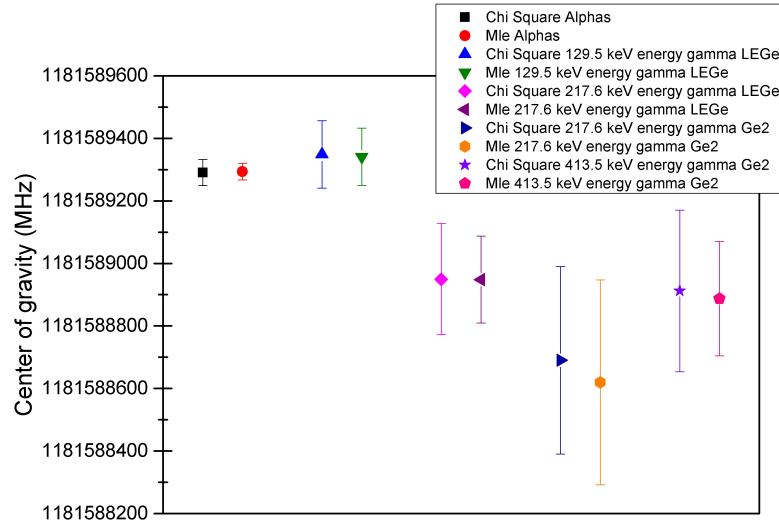


Figure 5.4: Comparison between the center of gravity for measurement 0366 obtained from the alpha and gamma HFS using both χ^2 -fitting (including χ^2_{red} -correction) and mle-fitting. The result from the alpha HFS is used for further analysis.

5.1.3 Fit results for ^{181}Hg

The spin I of ^{181}Hg is equal to $1/2$. Consequently the spectroscopic quadrupole moment Q_s is zero. Therefore (see section 2.2.2) the quadrupole interaction parameters (B) of the upper and lower level of the transition are fixed to zero in the fitting procedures. The magnetic dipole interaction parameter of the lower level (A_l) is also set to zero since the lower atomic level has $J = 0$ (see figure 3.3) and therefore doesn't generate a magnetic field at the origin of the nucleus.

Three measurements are available for ^{181}Hg : 181Hg_scan_0362, 181Hg_scan_0271_to_0274 and 181Hg_scan_0279_to_0280. In the rest of this chapter these measurements are referred to as measurement 0362, 4444 and 0000, respectively.

Measurement	0362	4444	0000
Profile	Voigt	Voigt	Voigt
χ_{red}^2	12.7	63.5	3.7
Center of gravity (MHz)	1181561331 (39)	1181561274 (55)	1181561146 (58)
Au (MHz)	15089 (49)	15077 (70)	14996 (74)

Table 5.2: Summary of fit results obtained for χ^2 -fitting of the HFS based on counting alpha particles for the three measurements of ^{181}Hg . The displayed errors are corrected for the χ_{red}^2 -value. The value for the center of gravity of measurement 0362 is used for the further analysis. The results of scans 4444 and 0000 are omitted in the determination of $\delta < r^2 >^{198\ 181}$ due to unreliable fit results.

HFS based on counting alpha particles

The alpha HFS for all three measurements were fitted with a Voigt profile, since the corresponding correlation plot for the χ^2 -fitting was reliable. The results of the Chi-Square fitting will be used in the further analysis and are summarized in table 5.2. The corresponding fits are displayed in figure 5.5.

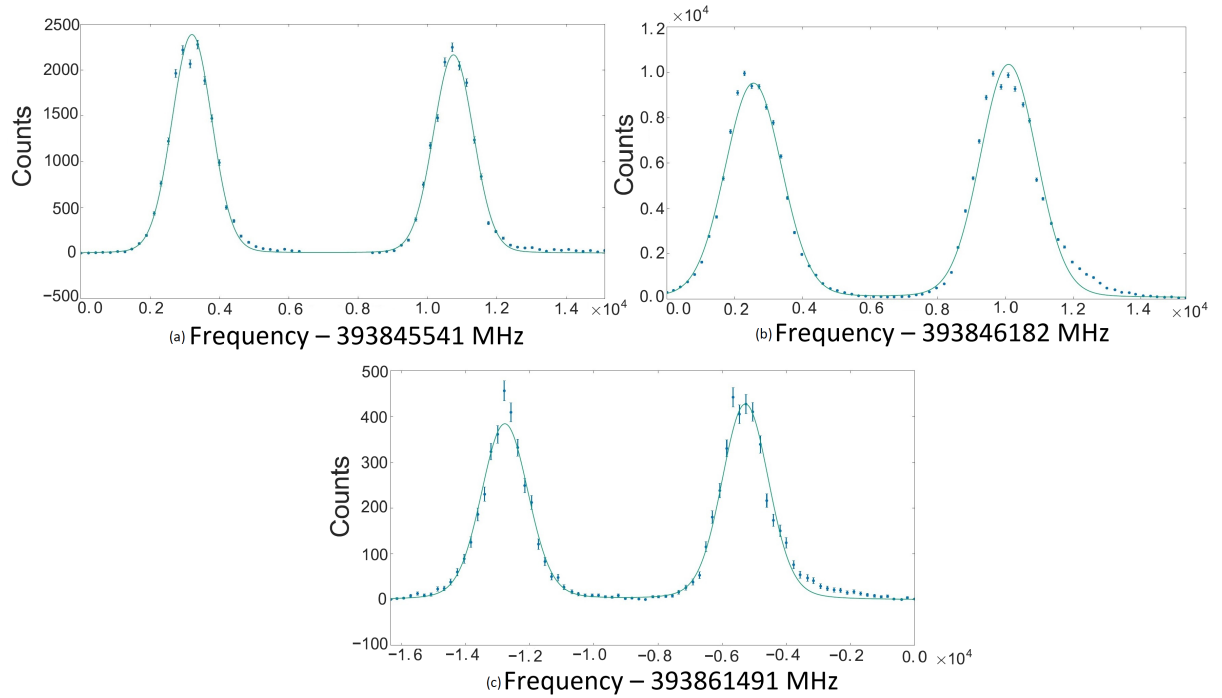


Figure 5.5: Fitted alpha HFS for the three measurements of ^{181}Hg . (a) Measurement 0362: the HFS is fitted with a Voigt profile ($\chi_{red}^2 = 12.7$). (b) Measurement 4444: a Voigt profile was used ($\chi_{red}^2 = 63.5$). (c) Measurement 0000: the HFS is fitted with a Voigt profile ($\chi_{red}^2 = 3.7$). The origin of the tail in the HFS displayed in (b) and (c) is unclear. The frequency difference with the first measured frequency is displayed. The results are summarized in table 5.2.

The values obtained with a χ^2 -fit and a mle-fit for the center of gravity and magnetic dipole moment of the upper level (A_u), after tripling, are shown in figure 5.6. The results of both fitting procedures are in good agreement for all three measurements. The results of A_u for the three measurements agree with the literature value $A_u = 14960$ (250) MHz [43]. The results for the center of gravity of measurement 0362 and 4444 are in agreement, while the value obtained from measurement 0000 deviates. However the HFS for measurements 4444 and 0000 exhibit a tail in one of the two resonance peaks (see figure 5.5). Due to this tail, the peaks of the HFS can not be properly fitted. The origin of this tail is unclear. This tail could have originated from the presence of contamination of ^{183}Hg , however this would cause tailing towards the other end of the peaks and is thus excluded. Measurement 0000 is taken immediately after measurement 4444 and is scanned in the reverse direction, while measurement 0362 is taken on another day. Therefore another possibility which cannot be excluded are fluctuations in the laser power at the end and beginning of measurement 4444 and 0000, respectively. The origin of this problem is subject of further investigation. Therefore it was decided to omit the results of those two measurements in the determination of $\delta < r^2 >^{198\ 181}$.

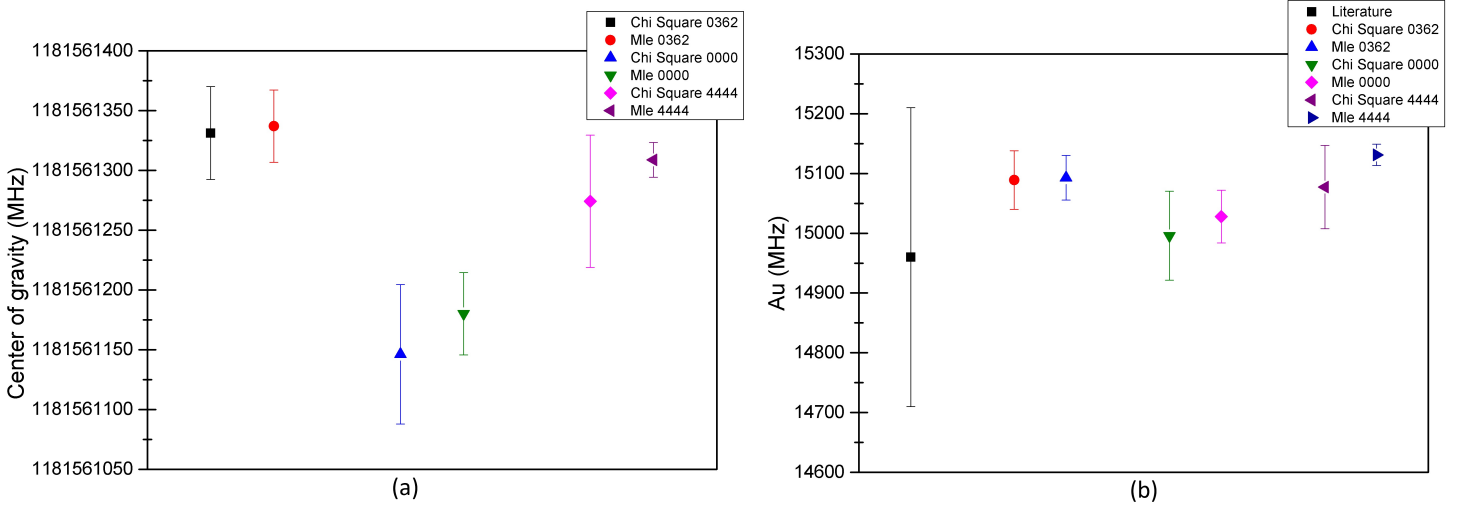


Figure 5.6: Results for (a) the center of gravity and (b) the magnetic dipole interaction parameter of the upper level (A_u) of ^{181}Hg , after tripling the fit values, for both χ^2 -fitting (including χ^2_{red} -correction) and mle-fitting derived from the alpha HFS for the three measurements of ^{181}Hg (0362, 4444 and 0000). Literature value is taken from [43].

HFS based on counting X-/ gamma rays

This section is discussed in appendix B since the gamma HFS for ^{181}Hg originate from measurements 4444 and 0000 which exhibit a tail and are therefore omitted in the determination of $\delta < r^2 >^{198\ 181}$.

5.1.4 Fit results for ^{198}Hg

With laser spectroscopy the mean square charge radius $\langle r^2 \rangle$ is determined with respect to a reference isotope. The used reference in this experiment is ^{198}Hg , which was frequently measured during the experiment in between the measurements of different Hg isotopes using both a Faraday cup and the Multi-reflection Time-Of-Flight set-up. Every measurement was done in both frequency directions in order to verify consistency.

^{198}Hg has $I = 0$ and consequently the magnetic dipole and quadrupole interaction parameters (A and B) of the upper and lower levels were fixed to zero in the fitting procedures, which were restricted to χ^2 -fits.

Faraday cup measurements

The first nine measurements of ^{198}Hg were performed using a Faraday cup (FC). The main disadvantage of a FC is that it measures the current of all the isotopes that were able to pass through the mass filter and there is no way to select just one isotope. This resulted in the presence of ^{197}Hg , both in the ground $^{197}\text{Hg}^g$ ($I = 1/2$) and isomeric state $^{197}\text{Hg}^m$ ($I = 13/2$), in the measurements of ^{198}Hg (see figure 5.7).

The magnetic dipole interaction parameter of the upper level (A_u) of both $^{197}\text{Hg}^g$ and $^{197}\text{Hg}^m$ as well as the quadrupole interaction parameter of the upper level (B_u) of $^{197}\text{Hg}^m$ were fixed to the literature values [43] during the fitting procedures. The other hyperfine parameters were set to zero due to spin considerations.

A Gaussian profile is used due to unreliable error results for a χ^2 -fit using a Voigt profile.

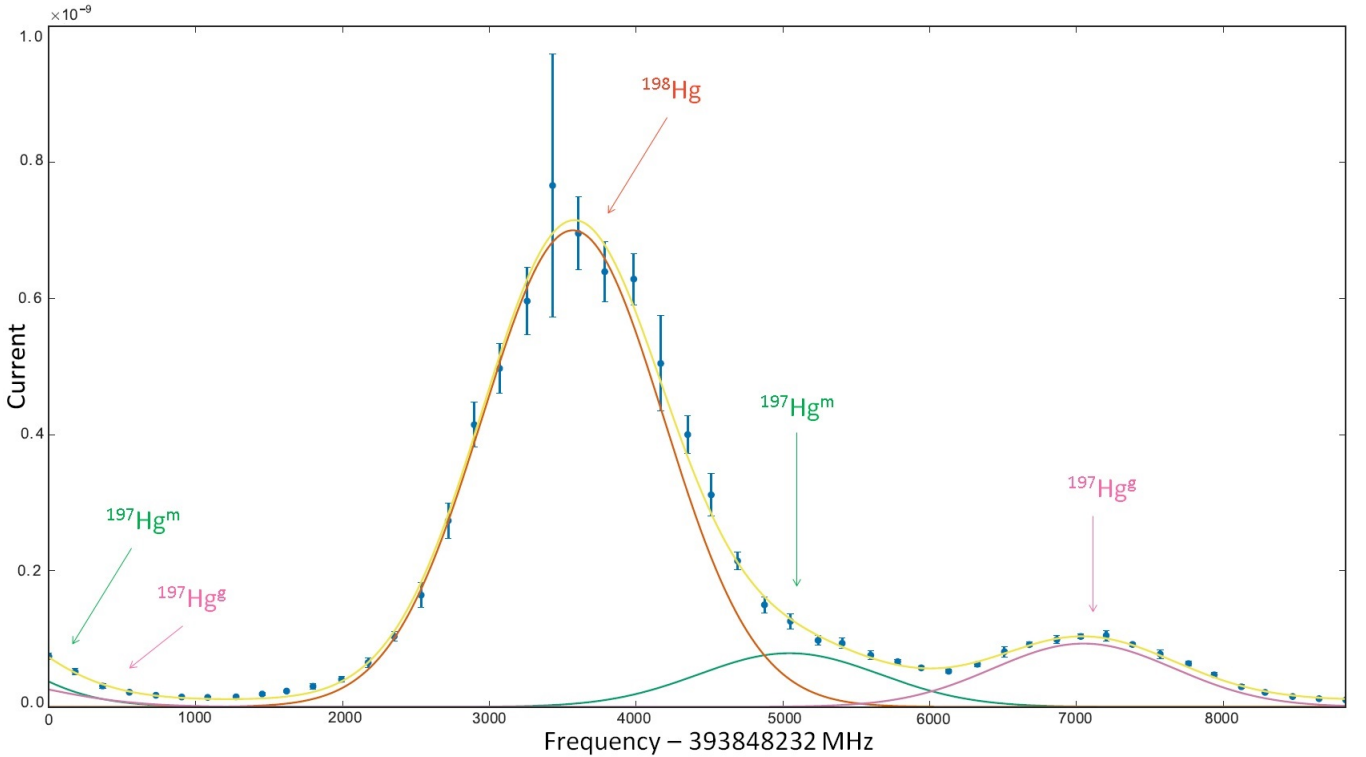


Figure 5.7: Fitted HFS measured with a FC for scan R5 of ^{198}Hg ($\chi^2_{red} = 1.7$). Contamination of $^{197}\text{Hg}^g$ and $^{197}\text{Hg}^m$ is taken into account.

Multi-reflection Time-Of-Flight measurements

In the eight last measurements of ^{198}Hg the Multi-reflection Time-Of-Flight (MR TOF) set-up was used.

The χ^2 -fit with a Voigt profile gave suspiciously large error bars for the full width at half maxima with a contribution of the Lorentzian profile (FWHML) close to zero. The computational time for a corresponding correlation plots was either excessive or there was a severe problem visible with the determination of FWHML. This problem arises from the limitation of FWHML to positive values, while the fit procedure needs the value of χ^2 for negative FWHML in order to determine the corresponding errors if FWHML is close to zero. Therefore it was decided to use a Gaussian profile instead.

The values of the center of gravity for ^{198}Hg , after tripling the fit results, are given in figure 5.8. It is not understood why there is such a large drift in the value of the center of gravity. Fluctuations in the wavelength readout can be the cause of these drifts. This problem is subject to further investigations.

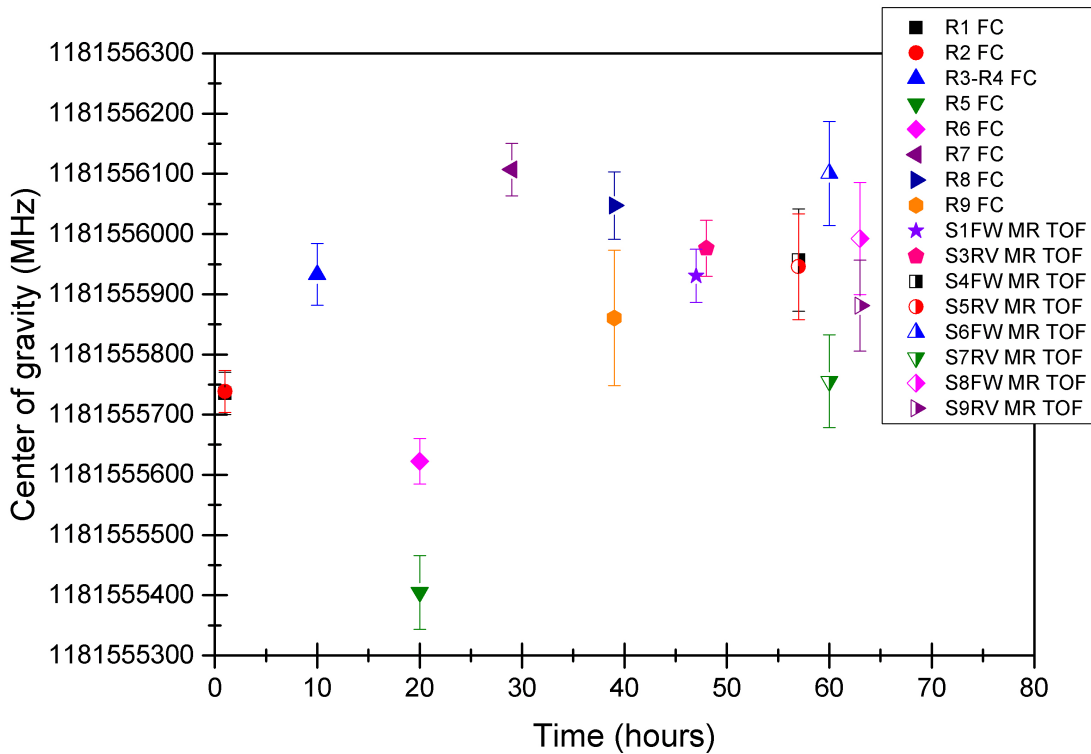


Figure 5.8: Values of the center of gravity for ^{198}Hg , after tripling the fit results. The errors are corrected for the χ^2_{red} -value. On the x-axis the time at which the measurement was carried out is given with the first measurement set to 1. When there are two scans shown on the same time, one scan is taken in the forward and the other in the reversed frequency order. Scan R3-R4 is the combination of both a forward and reverse measurement. A FC is used in the first nine measurements, while the MR TOF set-up is used in the last eight. It is not understood why there is such a large drift in the value of the center of gravity.

Averaging the reference values

The two scans of ^{182}Hg as well as scan 0362 of ^{181}Hg were measured one day after the last reference measurement of ^{198}Hg . Therefore the weighted average of all MR TOF scans is used as the reference COG value v^{198} in these measurements. This results in $v^{198} = 1181555941$ (23) {185} MHz. The first of the brackets gives the statistical error which is set equal to the weighted average error, the second indicates the systematic error which stems from the drift in the COG value. The systematic error is set equal to the maximal difference between the weighted average and the MR TOF data points, which is the difference with the COG obtained from scan S7RV.

The two other scans of ^{181}Hg were taken in between the reference measurements R3,R4 and R5,R6 performed with a FC. Therefore the weighted average between these results is used as the reference COG value v^{198} in measurements 0000 and 4444, which is equal to $v^{198} = 1181555725$ (24) {320} MHz. The systematic error is set equal to the difference between the weighted average and the COG result from scan R5.

The used reference value for each scan is summarized in table 5.3.

Scan	^{182}Hg : 0364 and 0366 ^{181}Hg : 0362	^{181}Hg : 0000 and 4444
Reference value v^{198} (MHz)	1181555941 (23) {185}	1181555725 (24) {320}

Table 5.3: Used reference value v^{198} for each measurement. Round brackets: the statistical error. Curly brackets: the systematic error which stems from the drift in the COG value.

5.1.5 General comment

An open question remains about how reliable the errors on the center of gravity are. As can be noted from tables 5.1 and 5.2, the COG is determined with an error of around a few tens of MHz (including χ^2_{red} -correction). Considering the facts that the width of the resonance peaks are around 4.5 GHz and that the distance between successive laser steps is around 600 MHz, this value seems to be too small. An error of around a few hundreds of MHz would be more reasonable. This is a general observed problem when fitting HFS and is still under investigation. An often used correction is to multiply the errors with a factor (e.g. 10) such that a reasonable value is obtained. In this work, no such correction on the errors was conducted. Future investigations will be performed to investigate the influence of this problem on the final results.

5.2 Determination of the isotope shift

In this section the isotope shifts (IS) $\delta v^{198,182}$ and $\delta v^{198,181}$ (table (5.4)) are determined. Only the results from the alpha HFS are used for this purpose (explained in section 5.1).

	$A = {}^{182}\text{Hg}$	$A = {}^{181}\text{Hg}$
$\delta v^{198,A}$ (MHz)	33377 (190)	5390 (191)

Table 5.4: IS values $\delta v^{198,182}$ and $\delta v^{198,181}$. The value between brackets is the combined error (statistical and systematic).

IS between ${}^{182}\text{Hg}$ and ${}^{198}\text{Hg}$

The two measurements of ${}^{182}\text{Hg}$ have the same reference value $v^{198} = 1181555941$ (23) {185} MHz. Therefore the weighted average of the COG values of both measurements is used to calculate the isotope shift. The combined COG value is $v^{182} = 1181589318$ (35) MHz in which the statistical error is the weighted average error. This value results in an isotope shift $\delta v^{198,182} = 33377$ (42) {185} MHz.

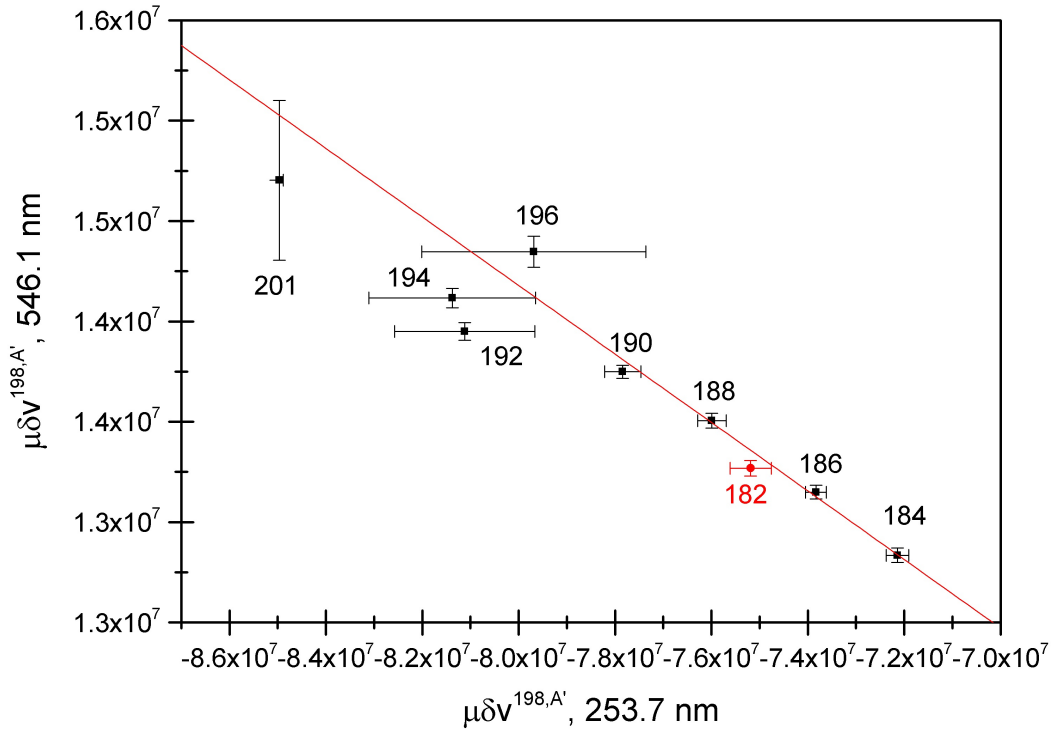


Figure 5.9: King plot of the IS in the 546.1 nm line versus those in the 253.7 nm line. ${}^{198}\text{Hg}$ is used as a reference isotope. The new data for ${}^{182}\text{Hg}$ coincides with the literature King plot line. Data taken from [14].

The IS of ${}^{182}\text{Hg}$ was not measured on the same atomic transition previously. Ulm et al. used the technique of collinear fast-beam laser spectroscopy [14] to measure the IS on the 546.1 nm transition. Therefore no direct comparison with literature can be made. However to verify consistency with earlier experiments, it can be checked if the new point for

^{182}Hg lies on the corresponding King plot line (explained in section 2.1.3). The slope of this line is $\frac{F_2^{AA'}}{F_1^{AA'}} = -0.1707$ (12) and the intercept can be determined from $K_{SMS,546.1} = 0.37$ (56) $m_e v_{546.1}$ and $K_{SMS,253.7} = 0$ (0.5) $m_e v_{253.7}$ with v the frequency of the transition[14]. The resulting King plot in the region of interest is shown in figure 5.9. The new point for ^{182}Hg coincides with the line.

IS between ^{181}Hg and ^{198}Hg

Due to the tailing in the HFS of scan 4444 and 0000, those two measurements were excluded from the determination of the isotope shift. Therefore, only the COG of scan 0362 is left, which is equal to $v^{181} = 1181561331$ (39) MHz. The reference value for this measurement is $v^{198} = 1181555941$ (23) {185} MHz. Therefore the isotope shift $\delta v^{198,181} = 5390$ (45) {185} MHz. This value is in agreement with the literature value $\delta v^{198,181} = 5560$ (200) MHz [43], as indicated in figure 5.10. The shown error includes both the statistical and systematic one.

Although the results from scans 4444 and 0000 are omitted in the analysis, it is interesting to compare the IS of all three measurements (figure 5.10). Since scans 4444 and 0000 are taken on another day than scan 0362, a different reference value is used (see table 5.3). After taken into account the reference measurements, all three results coincide. Therefore it can be concluded that the drifts in the COG values of ^{181}Hg (figure 5.6) have the same origin as the observed drifts in the reference measurement. This highlights the importance of taking several reference measurements during the experiment.

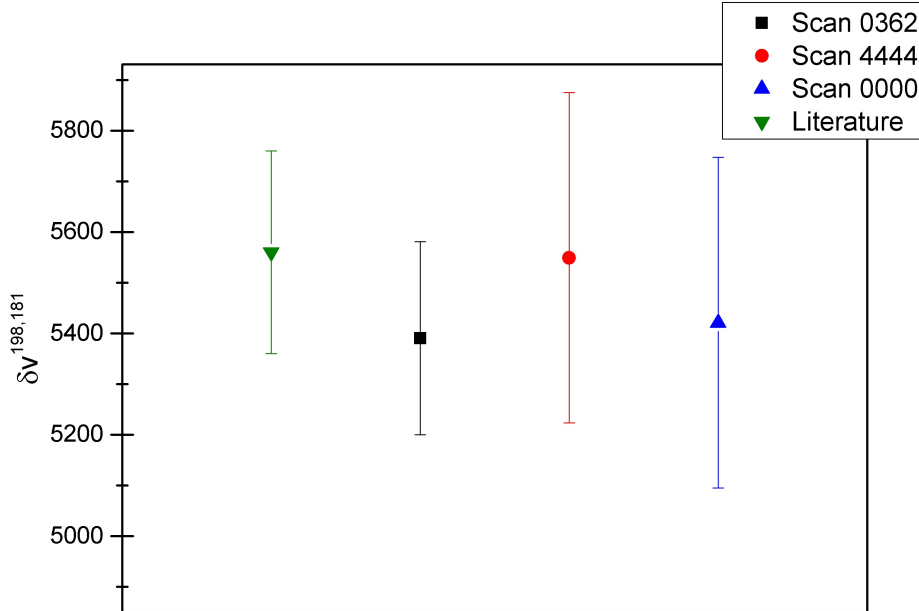


Figure 5.10: Comparison between the three values of the isotope shift obtained from the different measurements of ^{181}Hg and the literature value [43]. The three values coincide once the reference measurement is taken into account.

5.3 Determination of the change in mean square charge radius

$\delta \langle r^2 \rangle^{198\ A} \text{ (fm}^2\text{)}$	$A = ^{182}\text{Hg}$	$A = ^{181}\text{Hg}$
This work	-0.656 (0.004) {0.046}	-0.111 (0.004) {0.008}
Literature [14]	-0.651 (0.002) {0.047}	-0.114 (0.004) {0.009}

Table 5.5: Obtained values for the difference in mean square charge radii between $^{181,182}\text{Hg}$ and ^{198}Hg . Round brackets: purely experimental error. Curly brackets: systematic error resulting from the uncertainty of the F-factor and K_{SMS} .

The change in mean square charge radius $\delta \langle r^2 \rangle^{198\ 182}$ and $\delta \langle r^2 \rangle^{198\ 181}$ are determined from the IS values (table 5.4). Equation 2.8 is used with the atomic transition parameters of the 253.7 nm line: $K_{SMS} = 0(0.5)m_e v$ and $F^{198,204} = -55.36(7\%) \text{ GHz/fm}^2$ [14]. The slight A-dependence of the F-factor (equation 2.4) is not taken into account. This dependence is less than 1 % while the accuracy of the F-factor is not better than 7%. Therefore including the A-dependence of the F-factor seems to be redundant at the present level of accuracy [44]. The same approach is used for the odd-mass and even-mass Hg isotopes to extract the change in mean square charge radius from the isotope shift. In this work the approach of G. Fricke and K. Heilig [28] is used to relate $\lambda^{AA'}$ to $\delta \langle r^2 \rangle^{AA'}$, which states $\lambda^{198\ 181,182} = 0.927 \delta \langle r^2 \rangle^{198\ 181,182}$ (equation 2.7).

The errors are determined with standard error propagation. In this error calculation the uncertainty in the atomic parameters and the isotope shift value are taken into account, with a dominant contribution from the 7% uncertainty in the F-factor. The contribution of the uncertainty in the nuclear masses and the frequency of the transition are negligible.

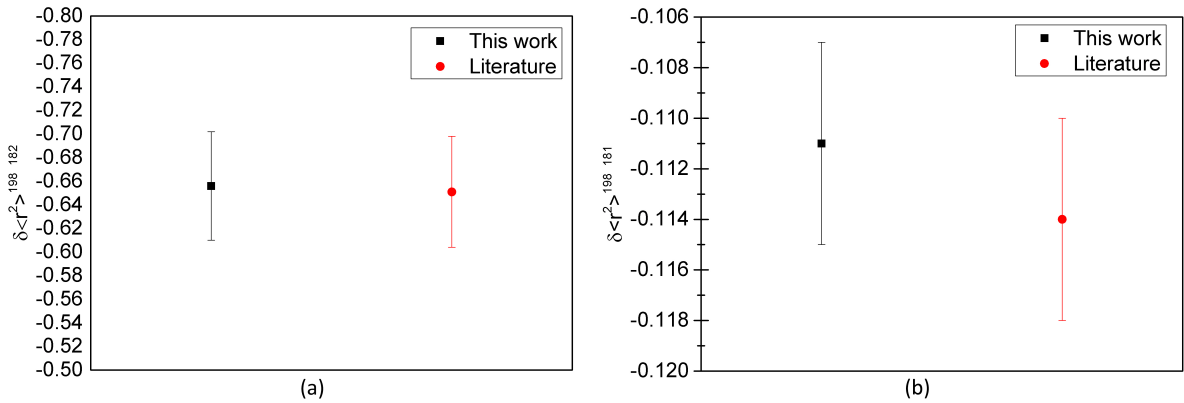


Figure 5.11: Comparison between the literature values [14] and the results in this work for (a) $\delta \langle r^2 \rangle^{198\ 182}$ (the errors include both the experimental and theoretical uncertainty) and (b) $\delta \langle r^2 \rangle^{198\ 181}$ (displayed errors are purely experimental).

The final results are presented in table 5.5 together with the literature values [14] and are compared in figure 5.11. Since the IS of ^{181}Hg was previously measured using the same transition, only the experimental errors are included in the comparison, whereas ^{182}Hg was previously measured using another transition and therefore both the experimental

and theoretical error have to be included. One can conclude that the results for $\delta < r^2 >^{198\ 181,182}$ are in agreement with the literature values. Due to the large theoretical uncertainties in $\delta < r^2 >^{198\ 182}$, more insight in the reliability of the in-source laser ionization technique is gained from the comparison using the King plot analysis (see figure 5.9). One can conclude that the measurements performed in April 2015 are reliable.

5.4 Determination of the rms deformation parameters

Parametrization form	$^{182}\text{Hg}: < \beta^2 >^{1/2}$	$^{181}\text{Hg}: < \beta^2 >^{1/2}$
Myers and Schmidt [5]	0.161	0.2771
Berdichevsky [6]	0.2091	0.3117

Table 5.6: Rms deformation parameters $< \beta^2 >^{1/2}$ for $^{181,182}\text{Hg}$

The difference in squared quadrupole deformation parameters $\delta < \beta^2 >^{198\ 181,182}$ can be roughly evaluated from $\delta < r^2 >^{198\ 181,182}$ (equation 2.11). The droplet model is used to express $\delta < r^2 >^{198\ 181,182}$ and $\overline{< r^2 >}_{\text{spherical}}$. Both Myers and Schmidt's [5] and Berdichevsky's [6] parametrization form (table 1.1) were applied to extract $\delta < \beta^2 >^{198\ 181,182}$. Since the rms deformation parameter $< \beta^2 >^{1/2}$ of ^{198}Hg has been calculated from the transition probability $B(E2)$ ($^{198}\text{Hg}: < \beta^2 >^{1/2} = 0.106$ (1) [14]), $< \beta^2 >^{1/2}$ of $^{181,182}\text{Hg}$ can be determined. The final results are presented in table 5.6.

The extracted deformation from the IS-data differs markedly for the two versions of the droplet model. Surprisingly, the results with the older version correspond better to deformations extracted from $B(E2)$ data (see [14]). Nevertheless some believe that Berdichevsky's version is more adequate and that the observed strange disagreement should be explained in a different way [44].

5.5 Preliminary results of isotopic chain behavior

In this work the consistency between the $\delta < r^2 >^{198\ 181,182}$ recently measured in ISOLDE and the literature values was verified. From this verification it can be concluded that the measurements performed in 2015 are reliable. All the other isotopes were analysed by the collaboration. In this section the preliminary results are presented.

The preliminary results of the changes in mean square charge radii $\delta < r^2 >$ in the isotopic chain $^{177-208}\text{Hg}$, relative to ^{198}Hg , is plotted in figure 5.12 as a function of mass number A . The regular and smooth variation of $\delta < r^2 >$ is restored for the neutron-deficient Hg isotopes behind ^{181}Hg , indicating that the ground state returns to a slightly deformed oblate state as was expected from theoretical considerations (see section 1.4.2 in chapter 1). From this observation, it can be concluded that $N = 82$ remains a magic number even if one approaches the proton dripline. Since the “parabolic-like” behavior of the excitation energy of the intruder state originates from the quadrupole-quadrupole interaction energy being maximal around mid-shell due to the largest number of valence nucleons to interact, if $N = 82$ indicates a closed shell structure.

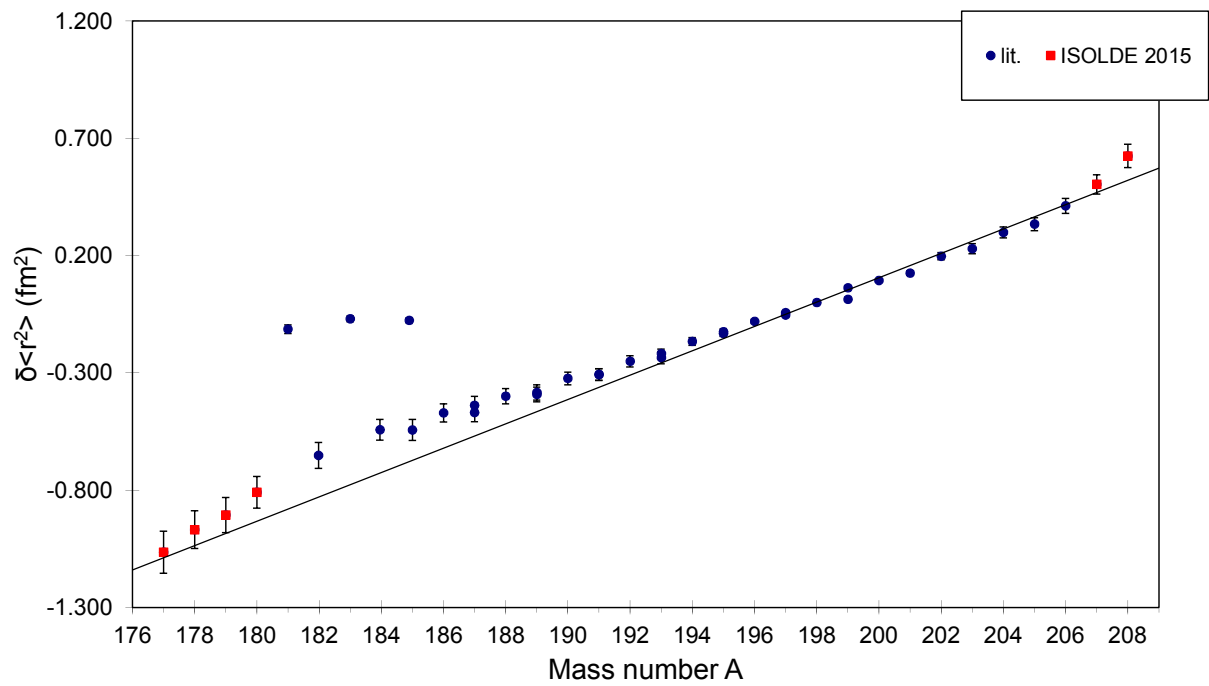


Figure 5.12: Preliminary results of the changes in mean square charge radii $\delta \langle r^2 \rangle$ in the isotopic chain $^{177-208}\text{Hg}$, with ^{198}Hg taken as a reference. Acknowledgment to Anatoly Barzakh. Blue: literature values [14]. Red: new data points measured in April 2015. The solid line is the prediction for the spherical droplet model, normalized to ^{198}Hg .

Chapter 6

Characterization studies at the IGLIS laser laboratory

A key element in the laser spectroscopy measurements, as described in the previous chapters, is the precise determination of the laser wavelength. However, during the past year a number of problems were identified in two wavelength meter models that are widely used at several laser laboratories, as explained in sections 6.2 and 6.3. A complete characterization and understanding of the wavelength meter performance becomes more important when the experimental precision increases. Problems were encountered with two different wavelength meters used for experiments at the LISOL facility [45]. As one of the wavelength meters was of the same type as the one used for the mercury laser spectroscopy measurements discussed in the previous chapters, a series of systematic measurements to characterize the performance of the wavelength meters were performed as part of this thesis work. The measurements were performed in the In-Gas Laser Ionization and Spectroscopy (IGLIS) laser laboratory recently commissioned in the Instituut voor Kern- en Stralingsfysica (IKS) at KU Leuven in the frame work of the Heavy Element Laser Ionization Spectroscopy (HELIOS) project. The objective of the HELIOS project is to develop a new, high-resolution, high-efficiency laser spectroscopy technique based on in-gas-jet resonance-ionization spectroscopy that will overcome the present experimental limitations that prevent the study of actinide and trans-actinide elements [46].

6.1 Laser laboratory

The IGLIS laboratory comprises a high repetition, high power laser system, an atomic beam unit, wavelength and power meters and a scanning Fabry-Pérot. The research goals of the IGLIS laboratory are to characterize and optimize the laser system and investigate new atomic transitions for future use in on-line applications. A photo of the laser laboratory is shown in figure 6.1 while a schematic layout of the lasers is shown in figure 6.2. In the following sections, the main components of the laser laboratory will be described.

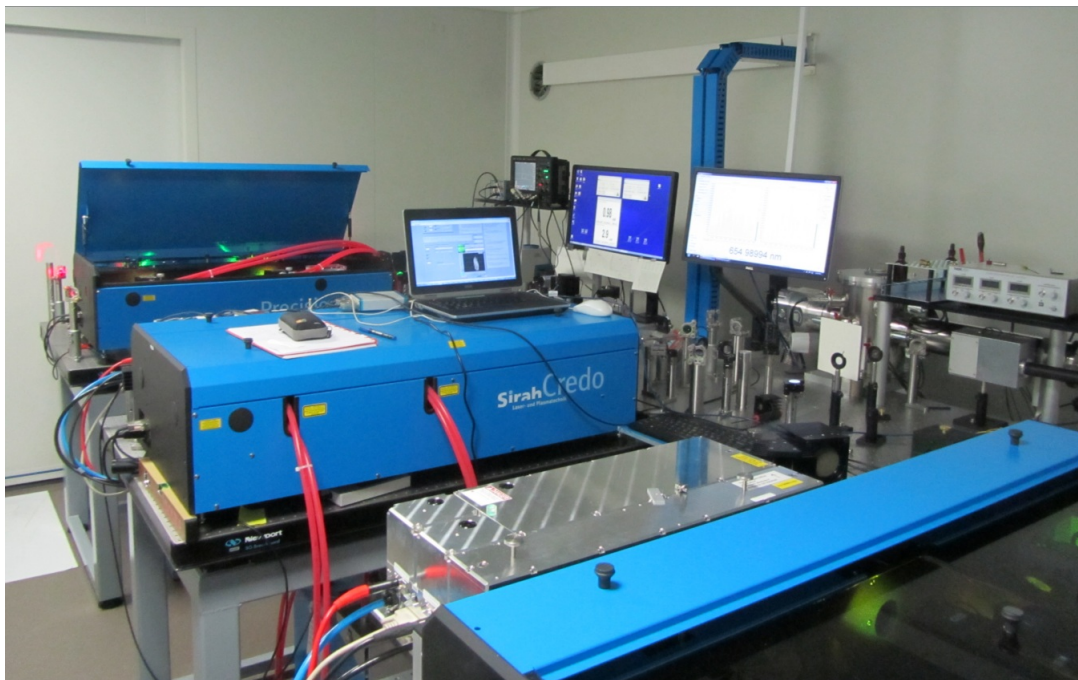


Figure 6.1: Photo of the laser laboratory. The Atomic Beam Unit (ABU) is located at the back next to the white computer screen. The lasers are placed on heavy metal tables. The laser closest to the wall is a first step narrow band laser. The laser next to it is a first step broadband laser followed by a second step broadband laser.

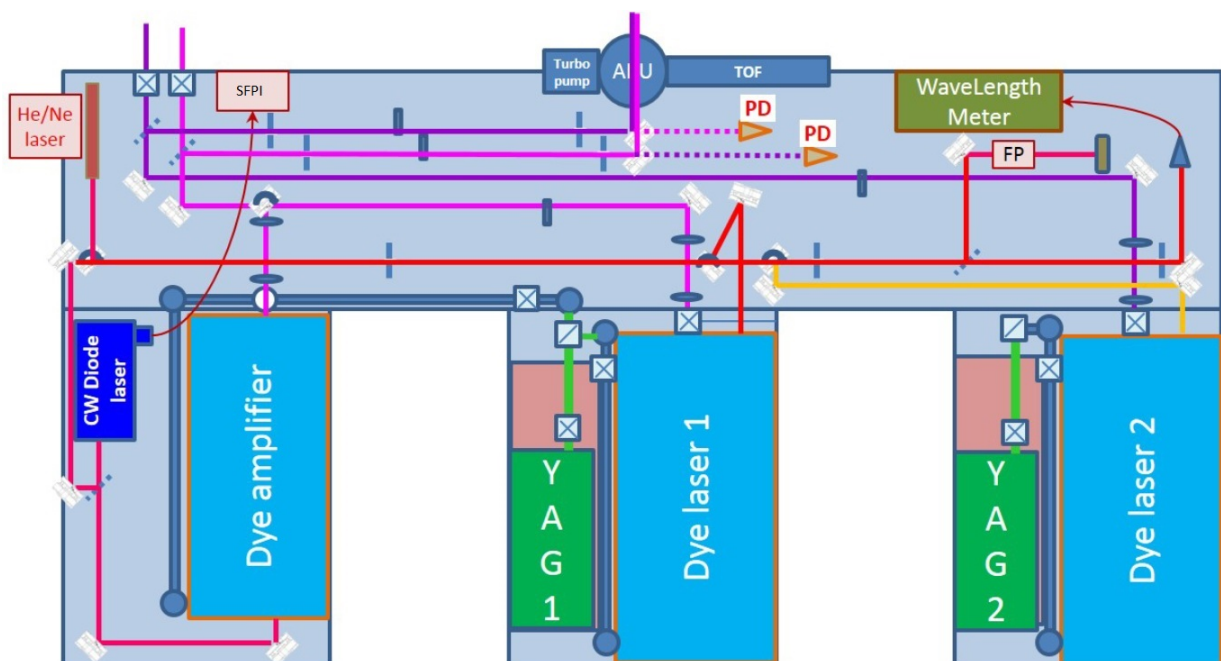


Figure 6.2: Schematic layout of the laser laboratory showing the positions of the ABU, the wavelength meters, the Scanning Fabry-Pérot Interferometer (SFPI) and the different lasers together with the optical path of the laser light. Courtesy of Yuri Kudryavtsev.

6.1.1 Atomic Beam Unit

An Atomic Beam Unit (ABU) is used to perform laser spectroscopy experiments on stable or long-lived isotopes.

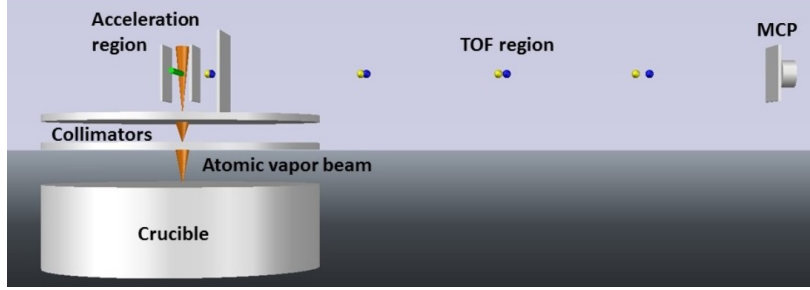


Figure 6.3: The Atomic Beam Unit. An atomic vapor is created by heating up copper wires inside the crucible. The resulting atomic vapor beam is collimated before it reaches the acceleration region where the atoms are ionized by a two-step laser ionization scheme. Once ionized the copper isotopes are accelerated towards a MCP ion detector. The signals of the different Cu isotopes can be distinguished because of their difference in arrival time at the MCP [47].

In the ABU of the IGLIS laboratory, shown in figure 6.3, small samples of natural copper are placed in a crucible. The crucible is resistively heated to a temperature of ~ 1250 K [47] and the copper samples evaporate. The opening in the crucible limits the divergence of the resulting atomic vapor which consists of $^{63,65}\text{Cu}$ (natural abundance : ^{63}Cu 69.15%, ^{65}Cu 30.85%). This atomic vapor beam is further collimated by two collimation plates before it reaches the ionization zone. The resulting low-divergence atomic beam is then ionized using a two-step laser ionization scheme, given in figure 6.4. The (first) excitation step requires laser light of 327.395 nm and the (second) ionization step is performed with a wavelength of 298.9 nm.

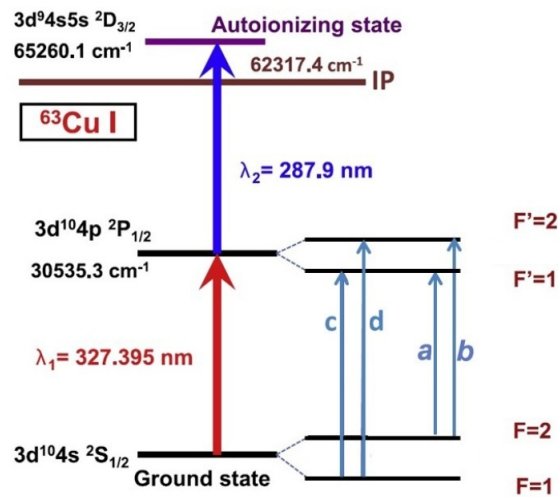


Figure 6.4: The two-step laser ionization scheme (not to scale) for ^{63}Cu used in the experiment [47].

The laser light is applied perpendicular to the atomic vapor beam. This geometry (together with the collimation) reduces the Doppler broadening. Once the atoms are ionized they are accelerated by means of a static electrical field towards a Multi-Channel Plate (MCP) detector. The difference in time-of-flight between ^{63}Cu and ^{65}Cu , due to their different mass-to-charge ratio, is used to distinguish the arrival time signals. This Time of Flight (TOF) Mass Separation can only be realized with pulsed lasers.

6.1.2 Lasers

A frequency-stabilized HeNe laser with a wavelength of 654.991026 nm (with an absolute accuracy of 10 MHz) is used to calibrate the wavelength meters and the Scanning Fabry-Pérot interferometer (SFPI).

Pulsed lasers are used in the HELIOS project because they have a higher power density and make it possible to saturate the transitions. In the laboratory a first step narrowband laser (bandwidth ~ 200 MHz), a first step broadband laser (bandwidth ~ 3000 MHz) and a second step broadband laser are available, shown in figures 6.1 and 6.2. These are a dye amplifier seeded with a diode laser and two dye lasers, respectively. A first step narrow band laser is necessary for in-gas-jet laser ionization spectroscopy experiments, while a broadband first step laser is sufficient for in-gas-cell experiments.

Nd:YAG lasers are used to pump the dyes. The dye amplifier is seeded with light from a single mode Continuous Wave (CW) diode laser which is amplified, pulsed and its output can be frequency doubled. This frequency doubling is necessary for the excitation of the Cu isotopes. The light of the CW diode laser is split in a beam going to the SFPI or a wavelength meter and a beam going to the dye amplifier.

6.1.3 Wavelength meters

Two wavelength meters are used in the laser laboratory: an ATOS (LM007) and a High Finesse WS-7, shown in figures 6.5a and 6.5b, respectively. Both wavelength meters have, according to the specifications of the producer, an absolute accuracy of 60 MHz around 632 nm.

Next to these two commercial wavelength meters, a Scanning Fabry-Pérot interferometer is available. The working principle is explained in the next section.

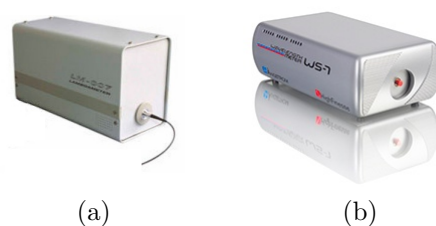


Figure 6.5: The available wavelength meters at the IGLIS laboratory. (a) ATOS. (b) WS-7.

Scanning Fabry-Pérot interferometer

A Scanning Fabry-Pérot interferometer (SFPI) is used to accurately determine the difference in wavelength between two nearby frequencies. The device consists of two partially

reflecting mirrors (see figure 6.6) forming a cavity. By applying a triangle voltage wave on the piezocrystals in between the two mirrors, the length of the cavity is periodically scanned during a measurement. A photodiode detector inside the SFPI measures the intensity of the light transmitted through the confocal SFPI cavity and its signal is observed on an oscilloscope, which displays the transmitted light intensity as a function of time and triggers at the rising part of the voltage.

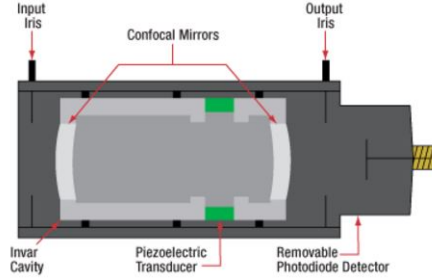


Figure 6.6: Schematic drawing of a SFPI [48].

For a certain distance between the two mirrors (L) wavelengths forming standing waves inside the SFPI cavity are transmitted. Those wavelengths obey the following boundary condition

$$L = \frac{n\lambda}{2} \quad \lambda = \frac{2L}{n} \quad f = \frac{c}{\lambda} = \frac{cn}{2L} \quad (6.1)$$

with n a natural number, λ the wavelength, f the frequency and c the speed of light.

The smallest difference between two frequencies (f_1, f_2) transmitted through the SFPI at the same distance L (and thus at the same time during a measurement) is equal to

$$f_1 - f_2 = \frac{c(n+1)}{2L} - \frac{cn}{2L} = \frac{c}{2L}. \quad (6.2)$$

This difference is known as the Free Spectral Range (FSR) of a SFPI, which is 4002 (18) MHz (see Appendix D) for the SFPI in the IGLIS laboratory. Note that the FSR does not change during the measurement since the change in the distance L is negligible. (The cavity length is around 40 mm, determined using equation 6.2, and is increased by roughly 2600 nm when measuring eight orders (explained in the rest of this section) of a wavelength around 655.98 nm.)

If the response of the crystal to the applied voltage would be every time exactly the same, the absolute wavelength could be determined (once the voltage-displacement relation was characterized) just by measuring the voltage difference between two successive constructive interferences of the light in the SFPI. Since this voltage difference could then be directly translated into a change of the distance between the two mirrors, being equal to $\lambda/2$, with λ the wavelength of the laser light. Unfortunately, SFPIs are very sensitive to the external conditions as e.g. changes in the room temperature or pressure. Thus the response of the piezocrystals to the applied voltage will be different for different measurements. Therefore a reference laser (e.g. a HeNe laser) has to be used to characterize the voltage-displacement response of the crystal and only differences between two nearby laser frequencies can be measured.

The principle of how a SFPI can be used to measure the difference in wavelength between two laser sources is explained by means of figure 6.7.

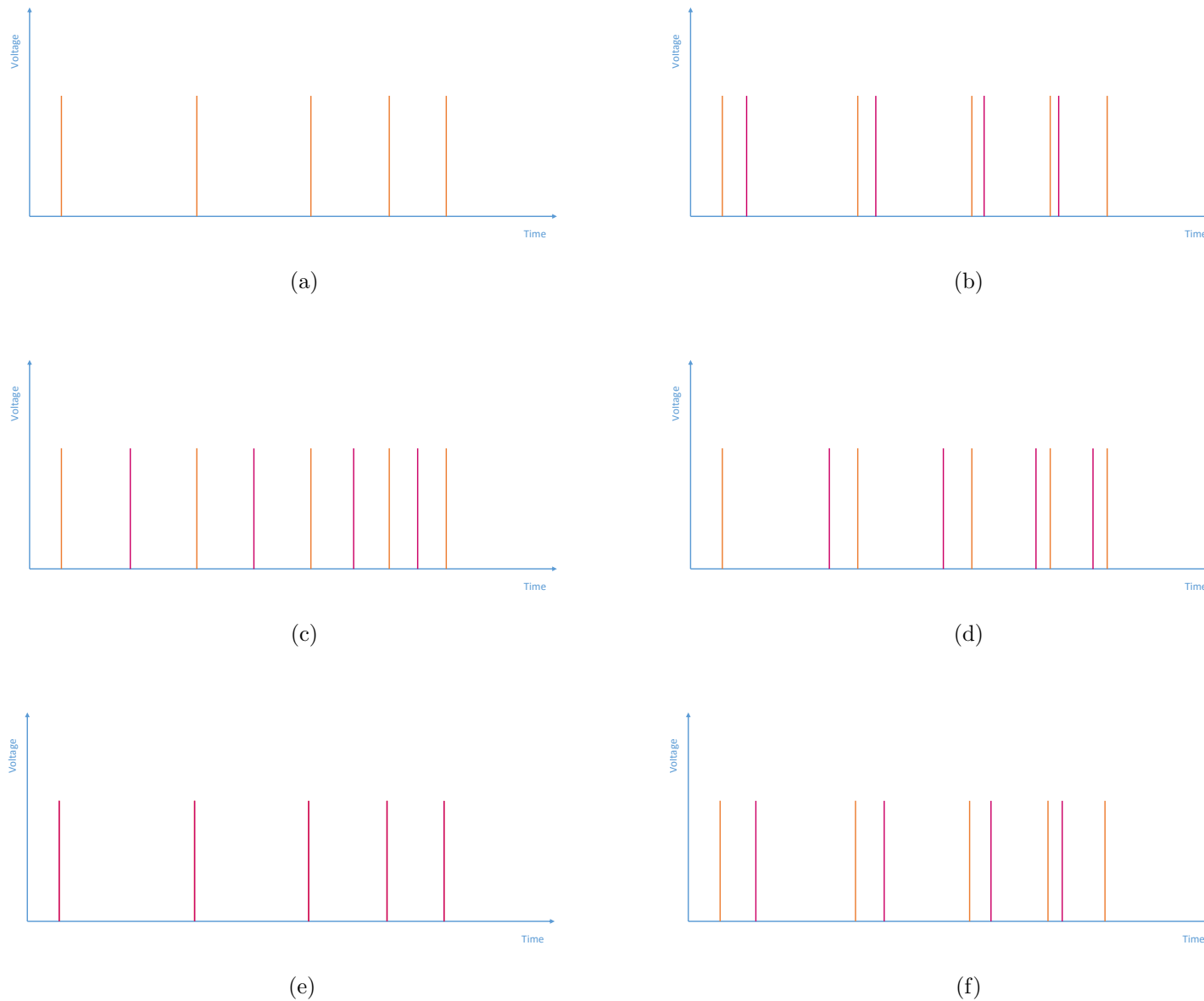


Figure 6.7: Illustrations used to explain the working principle of a SFPI. (a) The signal of a laser sent to a SFPI read by an oscilloscope. Every time an orange peak is visible, the light is transmitted through the SFPI. The difference in time between the peaks is not equal because of the non-linear voltage response of the piezocrystal. (b) Suppose that in (a) two lasers were connected to the SFPI with the same frequency. If the frequency of one of the two lasers is decreased, the peaks of this laser (pink) shift to the right assuming that the distance between the two mirrors is increasing. (c)-(d) The peaks keep shifting to the right if the frequency is further decreased. (e) Once the signals of the two lasers coincide, the frequency of the laser has decreased by one FSR. (f) The peaks keep shifting to the right if the frequency is further decreased.

When initially two lasers with exactly the same frequency are sent to a SFPI, the signal on the oscilloscope would look like figure 6.7a. The orange line indicates that the light is transmitted through the SFPI. After a certain time this signal reappears since the distance between the two mirrors is changing. The first peak is referred to as the first order, the second peak as the second order etc. The time difference between the peaks is not the same because of the non-linear voltage response of the piezocrystal. If now the frequency of one of the two lasers is decreased, the signal on the oscilloscope would look like figure 6.7b. The peaks of the second laser (pink line) have moved to the right, assuming that the distance between the mirrors is increasing during the measurement. If the frequency of this laser is decreased further, the peaks keep moving to the right (as in figures 6.7c and 6.7d) until the signals of the two lasers coincide (as in figure 6.7e). At this moment the frequency of the second laser has decreased by one FSR. If the frequency is decreased even more, the peaks keep shifting to the right (as illustrated in figure 6.7f) until the signals coincide again. At this moment the frequency of the second laser has decreased by two FSRs. Therefore, by fitting the time position of the orange peaks as a function of FSRs, the time position of the pink peaks can be converted to an energy in units of FSR i.e. of Hz.

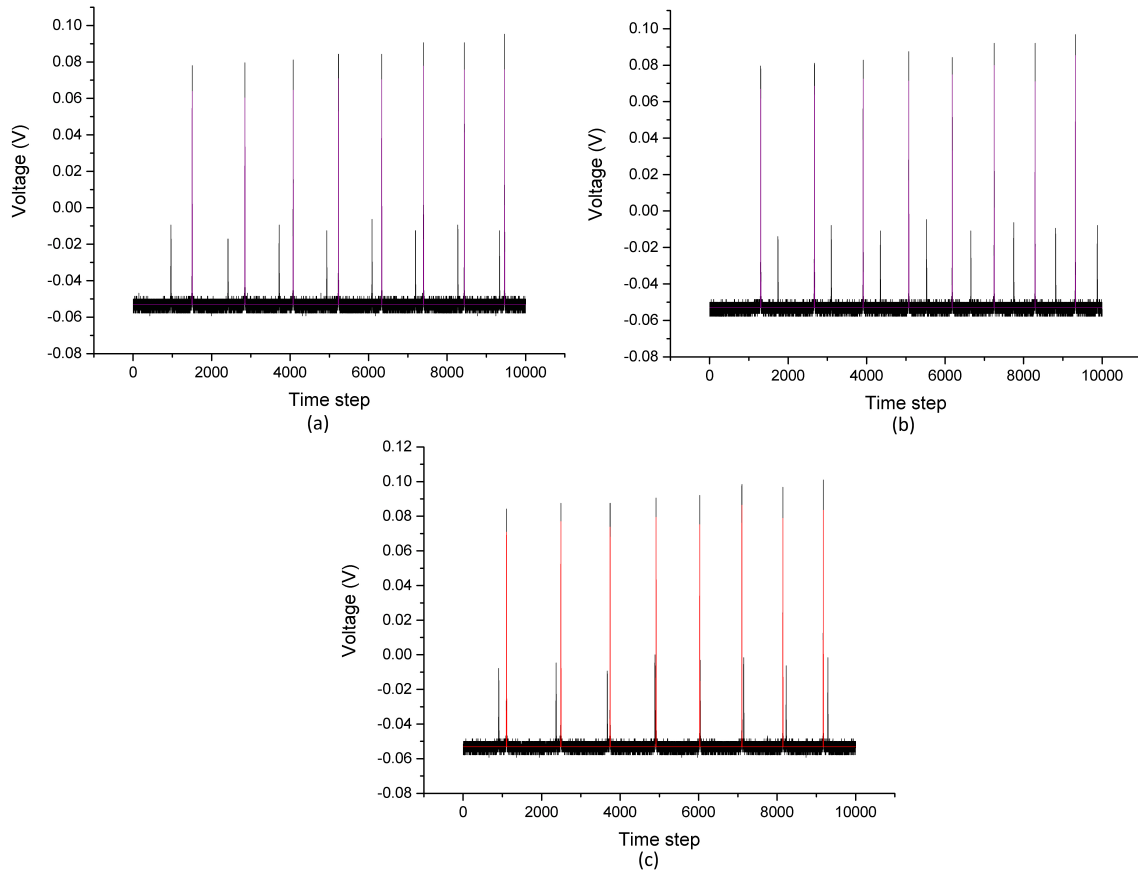


Figure 6.8: Examples of the oscilloscope readout during three measurements. The large peaks are the HeNe peaks, while the small peaks are the signals of the CW diode laser. The frequency of the CW diode laser has been increased in going from part (a) to (b) to (c) and the peaks shift to the left. In (c) the first diode peak has passed the first HeNe peak.

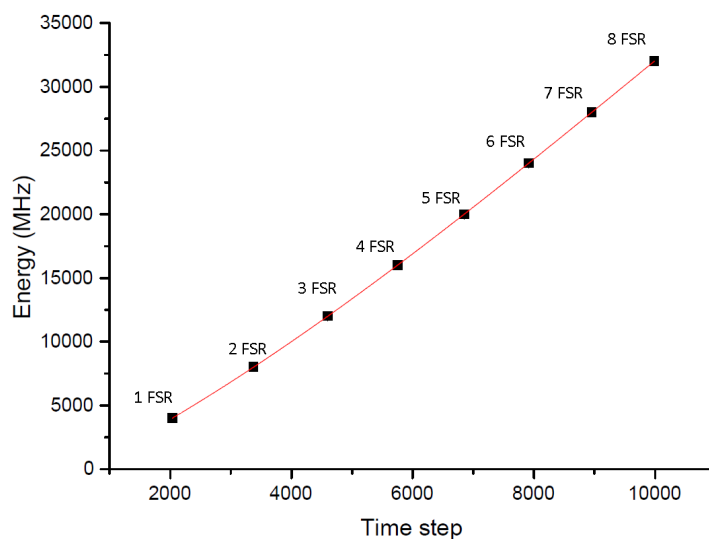


Figure 6.9: The time positions of the HeNe peaks, as in figure 6.8, are converted to an energy in units of FSR. The error in the time position is negligible, while the error in the energy is equal to 1 MHz. The curve is a fit using a cubic function to the data.

In order to investigate the linearity of the WS-7 wavelength meter used in spectroscopy studies on Cu (the wavelength meter used in the on-line measurements of mercury at ISOLDE is of the same type), the wavelength of the CW diode laser was scanned over the frequency range scanned in laser spectroscopy measurements on ^{63}Cu . A SFPI was used to accurately determine the difference in frequency between the first and currently measured wavelength of the CW diode. A HeNe laser is used for calibration and is also transmitted to the SFPI together with the CW diode laser. Examples of an oscilloscope readout during three measurements are given in figure 6.8.

The HeNe peaks are fitted with a gaussian function to determine their centroids. The time positions of these peaks are converted to an energy as demonstrated in figure 6.9. The error on the time position is negligible, while the error on the energy is estimated to be 1 MHz, conservatively taken 10% of the FWHM. The FWHM is around 10 MHz, determined by the finesse of the SFPI which is equal to 400 ($\text{FWHM} = \text{FSR}/\text{Finesse} = 4002/400$ MHz). The resulting graph is fitted with a polynomial. (A polynomial is a logical choice since the voltage response of a piezocrystal can be described by a Taylor expansion.) Such a graph obtained for the first measured CW diode wavelength was fitted with a linear, a quadratic, a cubic and a quartic equation. The linear function was included to demonstrate the non-linear behavior of the piezocrystal. Based on the analysis of the residuals (see figure 6.10), a cubic fit function was taken. With the aid of the obtained fit function ($E(t)$) the time positions of all the CW diode peaks can be converted to energy. This calibration was repeated for every measurement separately, since it was noticed that the fit function differed significantly from one scan to the next, as is clear from figure 6.11. In this figure the difference in time steps between successive HeNe peaks is plotted for every file that was measured during the wrong calibration (figure 6.11a) and the correct calibration (figure 6.11b) experiment (explained in section 6.3). This figure indicates that the SFPI is not stable.

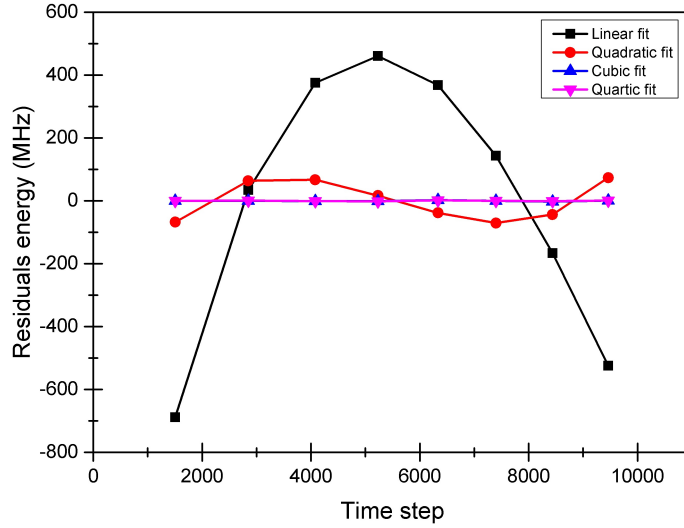


Figure 6.10: The residuals for a linear, a quadratic, a cubic and a quartic fit (not visible on the graph because the function overlaps with the cubic fit). The linear function was included to demonstrate the non-linear behavior of the piezocrystal.

The final energy value is given by the weighted average of all orders. In order to calculate the weighted average, the higher order peaks have to be translated towards the first order (i.e. the CW diode peak between the first and second HeNe peak) by subtracting the necessary multiples of one FSR. The weight given to each order is determined by the error from the fit. However since this error obtained with error propagation scales with the time position (e.g. the seventh order for the first measured CW diode wavelength has an error of 461 MHz compared to 87 MHz for the first order), those errors have to be rescaled. This rescaled error ϵ is given by

$$\epsilon = \text{error}_{\text{propagation}} * \frac{\text{Time position first order}}{\text{Time position present order}} \quad (6.3)$$

with $\text{error}_{\text{propagation}}$ the error resulting from the standard error propagation formula. The weighted standard deviation is used as error of the final energy values. This is possible since all orders are measured during one scan which takes a number of milliseconds.

The difference between the currently measured CW diode frequency (f_c) and the first measured frequency (f_1) can be determined by using the following formula

$$f_c - f_1 = (E(t_1) - E(t_c)) \times \frac{\lambda_{\text{HeNe}}}{\lambda_{\text{CW diode}}} + N_{\text{jumps}} \times \text{FSR} \quad (6.4)$$

with $E(t_1)$ the average energy of all CW diode peaks obtained with the aid of the fit function for the first measured frequency, $E(t_c)$ means the same but for the currently measured frequency. N_{jumps} is the number of times the CW diode peak has passed the first HeNe peak (see figure 6.8). When this happens one FSR has to be added to the average energy, since for every measured wavelength this average energy lies in between the first and second HeNe peak. λ_{HeNe} and $\lambda_{\text{CW diode}}$ are the wavelengths for the HeNe and CW diode laser, respectively. The factor $\lambda_{\text{HeNe}}/\lambda_{\text{CW diode}}$ has to be included because the calibration wavelength does not coincide with that of the CW light. Therefore the time between successive HeNe peaks is not the same as when a reference laser of the same

wavelength as the CW diode laser would be used. The sign of the first term in equation 6.4 is determined by the fact that the frequency is increasing, but the peaks are shifting to the left, as shown in figure 6.8.

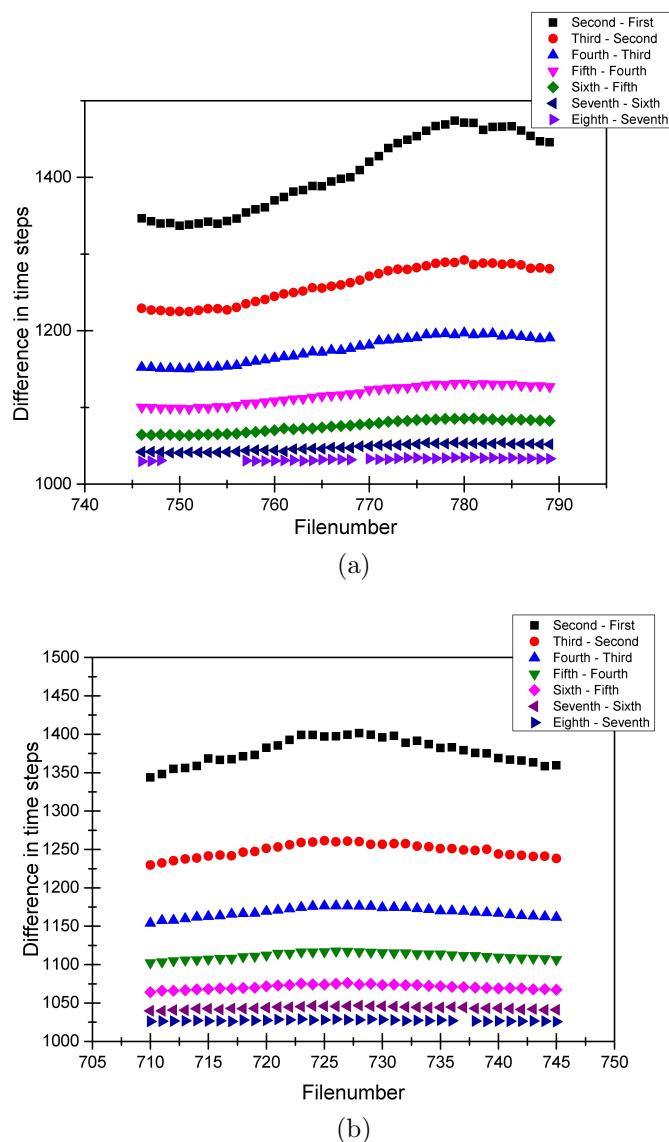


Figure 6.11: In this graph the difference in time steps between successive HeNe peaks in the SFPI readout is plotted for every file that was measured (a) during the wrong calibration and (b) the correct calibration experiment.

6.2 Discrepancies in the measurements of ^{215}Ac

In December 2014 the HFS of ^{215}Ac was measured using in-gas-jet laser spectroscopy at the Leuven Isotope Separator On-line (LISOL) set-up in Louvain-la-Neuve [45]. The laser ionization scheme is given in figure 6.12. The scan of the high energy singlet peak ($F_1=6 \rightarrow F_2=7$) with a wavelength around 438 nm was measured using both the WS-7 and ATOS wavelength meter. These wavelength meters were calibrated with a HeNe laser. An example of the obtained plots is shown in figure 6.13. The centroids are not in agreement. Ten measurements were performed with WS-7 and two measurements with ATOS. The average value for the peak positions is given in figure 6.14. The difference between the average peak positions for ATOS and WS-7 is around 750 (80) MHz. In order to understand and solve such a strong disagreement, it was decided to perform a systematic study to compare the wavelength readout between WS-7 and ATOS in different wavelength regions.

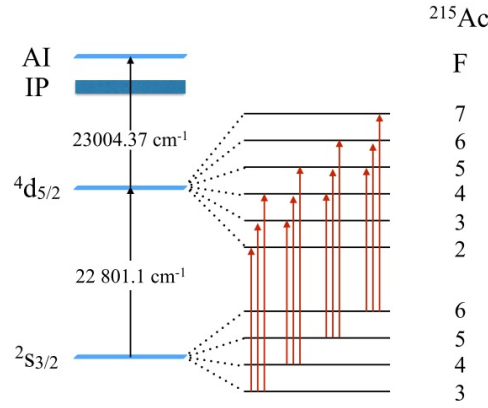


Figure 6.12: The two-step laser ionization scheme (not on scale) for ^{215}Ac used in the experiment [49].

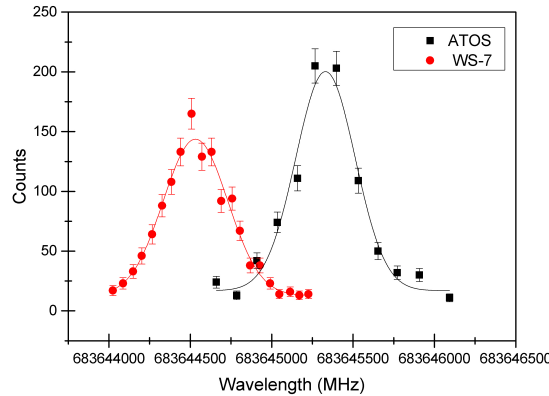


Figure 6.13: Example of the HFS for the high energy singlet peak of ^{215}Ac measured with WS-7 and ATOS. A clear mismatch between the peak positions is visible.

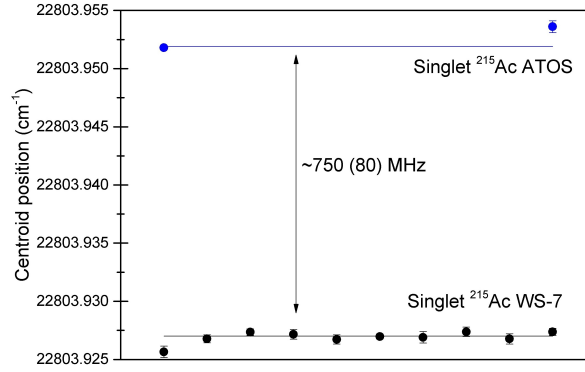


Figure 6.14: Average high energy singlet peak position of ^{215}Ac measured with WS-7 and ATOS. Courtesy of Rafael Ferrer.

6.2.1 Comparison of the wavelength readout between WS-7 and ATOS

The performance of the two wavelength meters (after calibration with the HeNe laser) was checked in different wavelength regions.

Two Toptica DL Pro lasers locked to the four D1 transitions ($S_{1/2} \rightarrow P_{1/2}$) of cesium and of potassium respectively were used to check the readout of both wavelength meters in the region around 895 nm and 770 nm. The results for cesium are displayed in figure 6.15a. The obtained values with WS-7 and ATOS are consistent for all four available wavelengths and match with the literature values. The results for potassium can be found in figure 6.15b. The readout of WS-7 is in agreement with the literature values for all four wavelengths, while ATOS agrees only for one wavelength. However taking into account the error of 60 MHz, the results obtained with WS-7 and ATOS are consistent in the region considered.

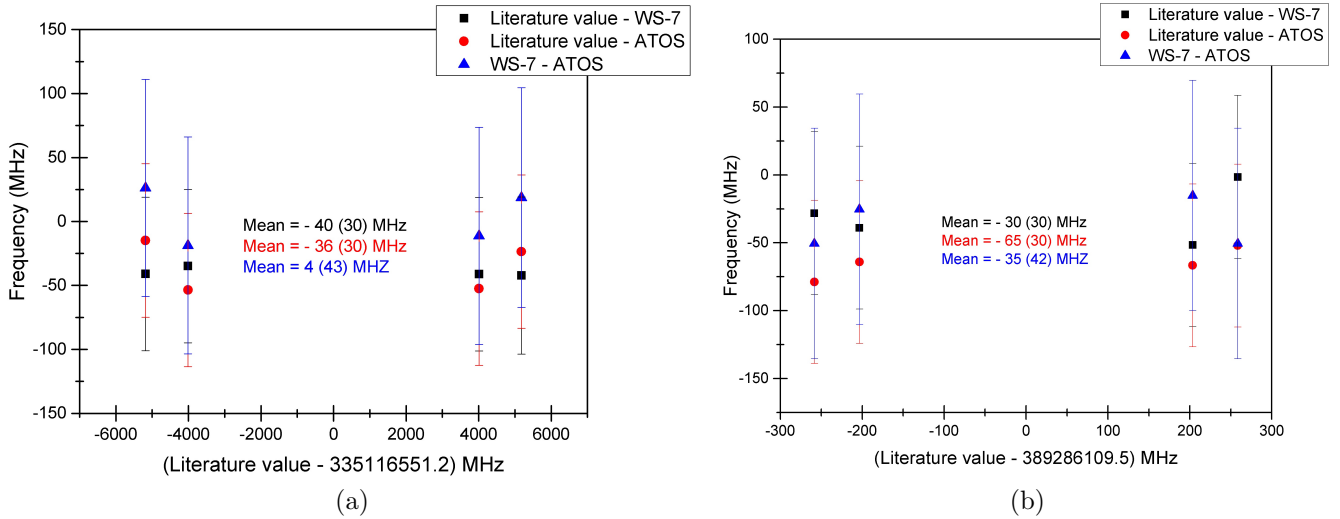


Figure 6.15: Two Toptica DL Pro lasers which were locked to the four D1 transitions of (a) cesium and (b) potassium are used to compare the readout of both wavelength meters in the region around 895 nm and 770 nm, respectively. Red: difference between the literature value and the readout of ATOS. Black: difference between the literature value and the readout of WS-7. Blue: Difference between the values measured by WS-7 and ATOS.

The fundamental and second harmonic light from the CW diode laser is used to compare the behavior around 655 nm and 327 nm. Therefore the light from the fundamental is sent to one wavelength meter while the second harmonic light is sent to the other one. By swapping the optical fibers between the wavelength meters the difference in readout in the fundamental and second harmonic light can be measured. The results for different measurement periods are grouped together in figure 6.16a. The difference in the fundamental $f_{WS-7} - f_{ATOS} = -49$ (17) MHz and the difference in the second harmonic $f_{WS-7} - f_{ATOS} = -1359$ (19) MHz with f_{WS-7} the frequency measured by WS-7 and f_{ATOS} by ATOS. The difference in readout of the fundamental is negligible while the difference in the second harmonic is unacceptably large. However from these results only it can not be concluded which wavelength meter is measuring the Ac data more correctly. Therefore the linearity of both detectors was checked using the fact that the detector which performs well in the second harmonic region has a difference between the frequency of the fundamental and the second harmonic divided by two ($f_{Fun} - \frac{f_{SH}}{2}$) close to zero. The results for the linearity are given in figure 6.16b. WS-7 measures the second harmonic fairly well with a difference $f_{Fun} - \frac{f_{SH}}{2} = 69$ (15) MHz, while $f_{Fun} - \frac{f_{SH}}{2} = -557$ (15) MHz for ATOS. This means that the wavelength readout of the second harmonic by ATOS is shifted towards the blue.

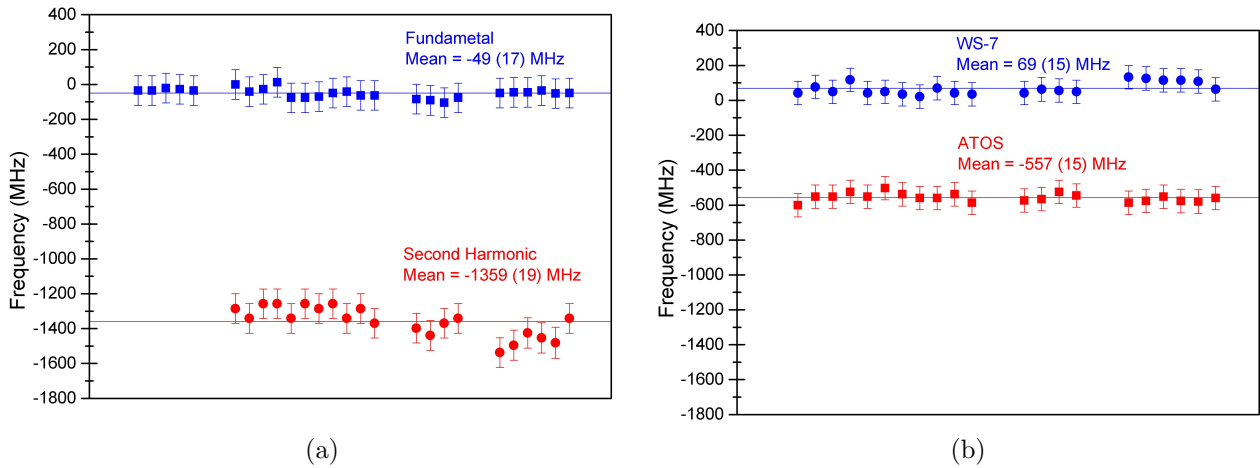


Figure 6.16: (a) Comparison in readout of the fundamental (≈ 655 nm) and second harmonic (≈ 327 nm) light generated by the CW diode laser. (b) Linearity of both wavelength meters.

All the results for $f_{WS-7} - f_{ATOS}$ measured in different wavelength regions are summarized in figure 6.17. In this graph also the difference measured for Ac around 438 nm, the calibration point and the difference previously measured using a diode laser around 400 nm (borrowed from the LARISSA lab in Mainz) are added. From this graph it can be concluded that ATOS and WS-7 give a comparable value in the region between 632 nm and 885 nm. However the results deviate significantly for shorter wavelengths. A possible solution to overcome this problem would be the use of a calibration laser as close as possible to the wavelength to be measured.

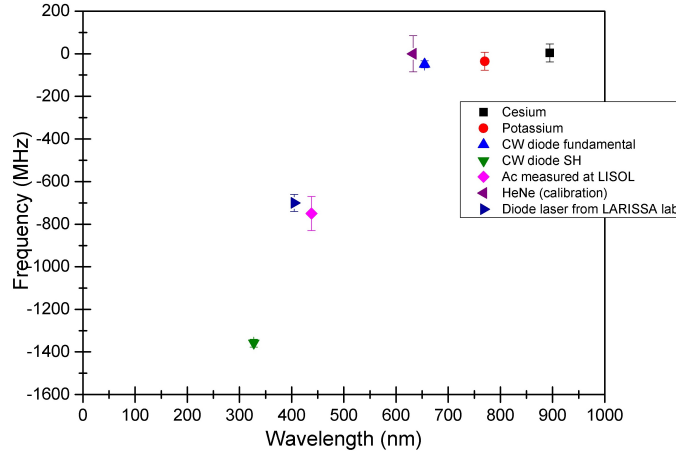


Figure 6.17: The difference in wavelength readout was checked in the region between 327nm and 885 nm. Also the shift observed in the Ac measurement and the calibration point are added. ATOS and WS-7 give a comparable value in the region between 632 nm and 885 nm, while the results deviate significantly for shorter wavelengths.

All the previous results were sent to the manufacturer of ATOS. They sent us a new program with a recalibration of the interferometers and the measurements with the CW diode light were repeated. The results are given in graph 6.18. The difference in the second harmonic light $f_{WS-7} - f_{ATOS} = 127$ (19) MHz is now much smaller and also the linearity of ATOS $f_{Fun} - \frac{f_{SH}}{2} = 135$ (12) MHz has improved. With the new program the readout of ATOS is red shifted compared with the one of WS-7. An extrapolation between the calibration point and the second harmonic gives an rough estimate of the shift between the value obtained with WS-7 and ATOS if an Ac measurement were to be performed with the new program. The extrapolation suggests a shift around 81 (67) MHz. It can be concluded that the problem is solved.

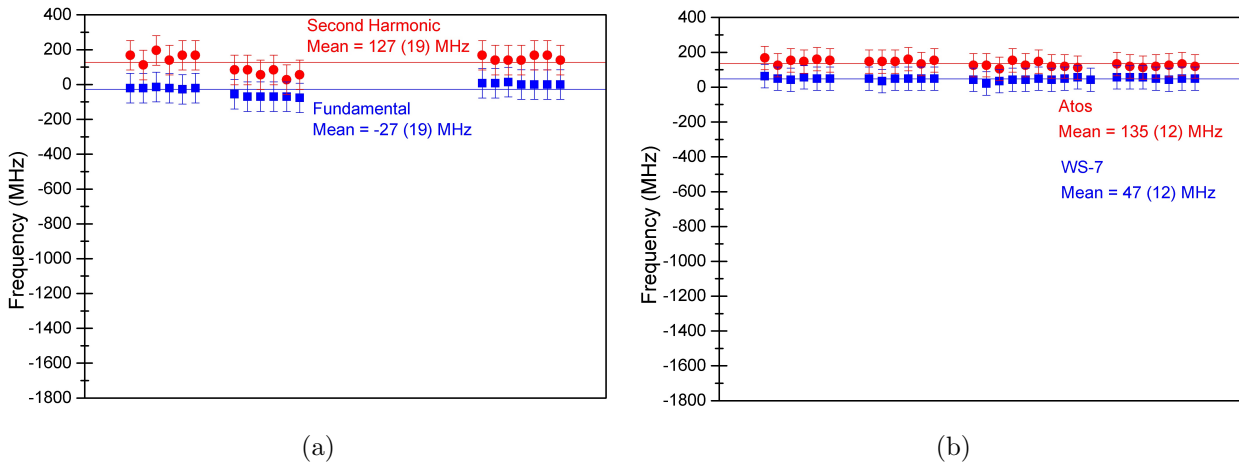


Figure 6.18: (a) Comparison in readout of the fundamental (≈ 655 nm) and second harmonic (≈ 327 nm) light generated by the CW diode laser and (b) the linearity of both wavelength meters with the new calibration of the ATOS interferometers.

6.3 Discrepancies in the measurements of ^{63}Cu

Last year laser spectroscopy experiments of ^{63}Cu were performed using the ABU. The Cu atoms were ionized by applying the two-step laser ionization scheme shown in figure 6.4. The second harmonic of the CW diode laser was used for the first excitation step. The wavelength of the fundamental was changed and measured by the WS-7 wavelength meter. The resulting HFS is given in figure 6.19.

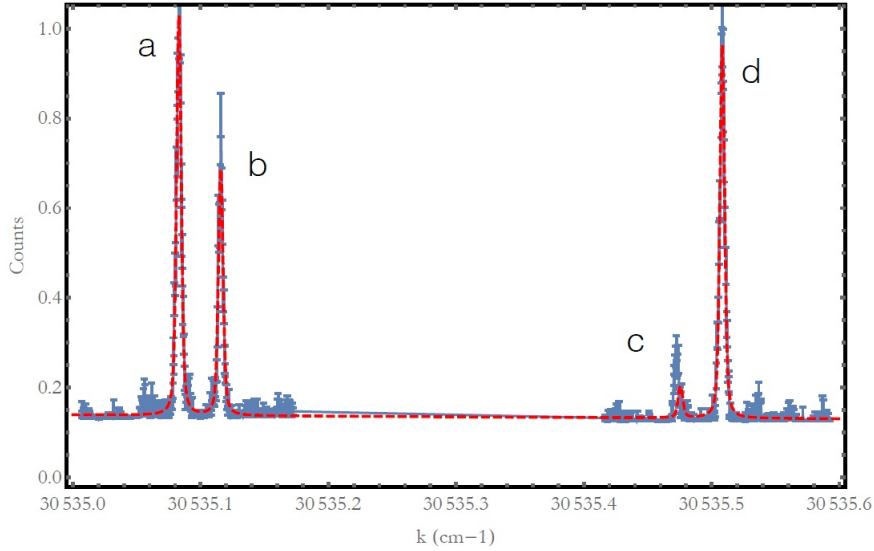


Figure 6.19: HFS of ^{63}Cu with the transitions labeled as in figure 6.4. The red dashed curve is a fit through the data assuming fixed hyperfine parameters. Courtesy of Sebastian Reader. In this graph some side bands are present. The discussion of these is outside the scope of this thesis [50].

This peak positions for the fitted HFS were compared with the literature values. It is clear from figure 6.4 that the distance between the peaks a and b ($|ab|$) must be equal to the distance between the peaks c and d ($|cd|$), since this distance is equal to the hyperfine splitting of the excited state ($^2P_{1/2}$). However, this is not the case for the measured HFS in figure 6.19. The red dashed curve is a fit through the data assuming fixed hyperfine parameters. This fit does not fit peak c properly. The literature value for the hyperfine splitting of the excited state ($^2P_{1/2}$) is 1013 (20) MHz [51] while the measured distance $|ab| = 979.48$ (17) MHz and $|cd| = 1075.14$ (17) MHz. These values result from fitting figure 6.19 without restrictions on the hyperfine parameters. After the measurements, a mistake was identified in the calibration file of the WS-7 meter. To verify that this was the cause of the observed problem, it was decided to test the WS-7 with the SFPI, as explained in the next section.

6.3.1 Comparison between WS-7 and SFPI

In order to investigate the problems in the HFS of ^{63}Cu the wavelength of the CW diode laser was scanned over the frequency range of the whole HFS. A SFPI was used to accurately determine the frequency change of the CW diode laser. At the same time the frequency was also measured with the WS-7 wavelength meter. Once calibrated with the

correct calibration ($\lambda_{\text{HeNe}} = 632.991026 \text{ nm}$) and once with a wrong calibration ($\lambda_{\text{HeNe}} = 632.99111 \text{ nm}$). This wrong calibration is the one that was accidentally used to measure the HFS as displayed in figure 6.19. The wavelength of the CW diode laser was scanned in steps of 0.00040 nm as measured with the WS-7. The difference in frequency between the chosen laser steps measured by the WS-7 and that measured by the SFPI is compared for the whole scanning range. In this way, the performance in terms of linearity of the WS-7 wavelength meter could be investigated.

The method used to determine energy differences with a SFPI was explained in section 6.1.3. The error on the wavelength readout of WS-7 only includes the statistical error, since the systematic error on the absolute value cancels out by taking differences. The statistical error was determined by observing the long-term fluctuations in the readout of the wavemeter. The fluctuations at the moment of recording the data were at most 4 MHz . Therefore 4 MHz was used for the error on the wavelength readout by the WS-7. The error on the differences are calculated by standard error propagation.

The first two orders are not reliable, since the voltage increases only in a stable linear way near the end of the ramp. Therefore those two orders were excluded from the analysis.

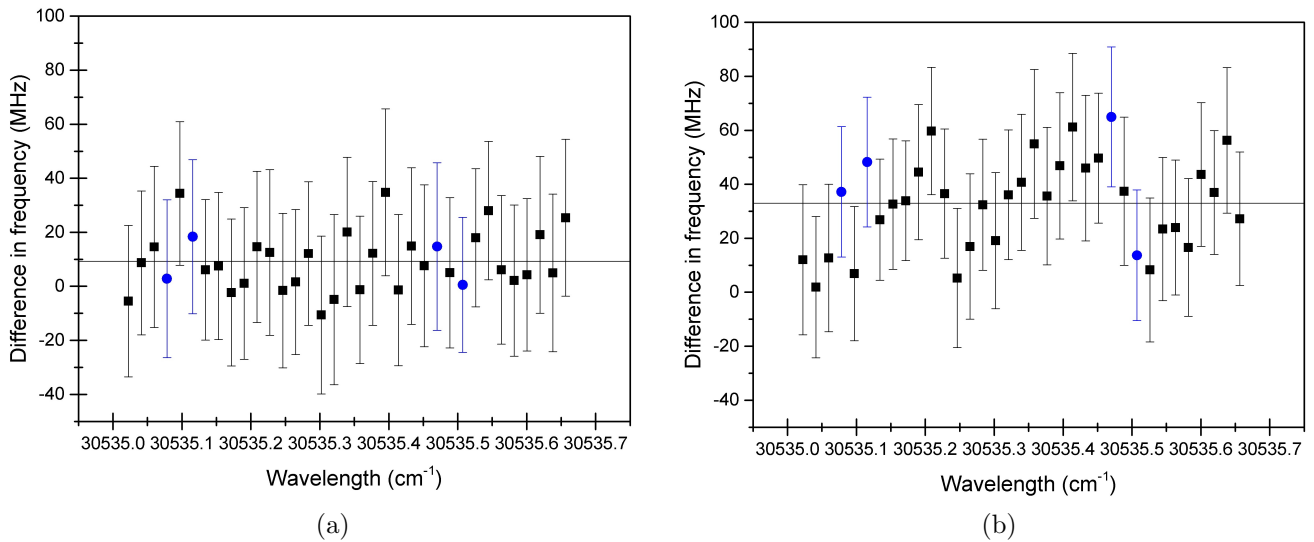


Figure 6.20: The wavelength of the CW diode laser was lowered in steps of 0.00040 nm as indicated by WS-7. The difference in frequency between the currently and first measured laser frequency with the SFPI is compared to the difference obtained with WS-7. The blue points represent the peak positions in the HFS of ^{63}Cu , shown in figure 6.19. The black line indicates the weighted average value. Only the statistical error is shown. The contribution of the systematic error on the FSR is 18 MHz . (a) Results when WS-7 was calibrated with the correct HeNe wavelength. (b) WS-7 calibrated with the wrong HeNe wavelength.

The final results for the difference in frequency between the chosen laser steps measured by the WS-7 and by the SFPI are displayed in figure 6.20. Only the statistical error is shown. The systematic error originating from the error on the FSR is 18 MHz , as explained in Appendix D. Part (a) shows the results for the correct calibration and part (b) for the wrong calibration, after doubling the values. Note that the results for the

correct calibration are normally distributed around the mean value as is expected from pure statistical fluctuations, while in the wrong calibration a systematic shift (a saw-tooth pattern) is evident that affects certain frequency points, among those is the peak c of the ^{63}Cu atomic spectrum (see figure 6.19). It can be concluded that the distance $|ab|$ should be equal to $|cd|$ for the correct calibration, while this is not the case for the wrong calibration as while peaks a and b are shifted with the same amount, peaks c and d are not and a difference around 60 MHz is observed.

6.4 Summary

During the past year a number of problems were identified in two wavelength meter models (i.e. WS-7 and ATOS) that are widely used at several laser laboratories; viz. there were problems of consistency between both wavelength meters in the measurement of the high energy singlet peak of ^{215}Ac and the hyperfine spectrum of ^{63}Cu could not be reproduced when using WS-7. As one of the wavelength meters (WS-7) was of the same type as the one used for the mercury laser spectroscopy measurements, a series of systematic measurements to characterize the performance of the wavelength meters were performed. The problems could be reduced to: a wrong calibration of the ATOS interferometers which was solved by the manufacturer and a wrong calibration of the WS-7 wavelength meter. The hyperfine spectra of the mercury isotopes were measured with the correct calibration file.

Chapter 7

Conclusion and outlook

The hyperfine spectra of 15 different mercury isotopes ranging from ^{177}Hg to ^{208}Hg were measured using in-source laser ionization spectroscopy at the ISOLDE facility in CERN in April 2015. The mean square charge radius of the mercury isotopes exhibits a large odd-even staggering for the neutron-deficient mercury isotopes, which was interpreted to be a manifestation of shape coexistence. One of the main goals of this experiment was to extend the currently existing knowledge of the mean square charge radius behavior down towards the more neutron-deficient isotopes and verify if the mercury isotopes become spherical again or whether the more neutron-deficient nuclei become even more deformed. In this thesis work, a subset of the data obtained in this experiment (viz. $^{181,182}\text{Hg}$) was analyzed to verify the reliability of the new method.

The mercury isotopes were produced via a spallation reaction induced by a collision of accelerated protons with a molten lead target. The mercury isotopes were ionized using a three-step laser ionization process after which they were mass separated and sent to a detection system. The reference nucleus, ^{198}Hg , was detected using a Faraday cup or the Multi-reflection Time-Of-Flight set-up with a multichannel-plate ion detector, while the $^{181,182}\text{Hg}$ isotopes were sent to the Windmill system. In this set-up the isotopes are implanted in carbon foils and the alpha decay was detected using silicon detectors. The aim of the experiment was to construct the hyperfine spectra of which the isotope shift and change in mean square charge radius can be determined.

In this work the consistency between the $\delta < r^2 >^{198\ 181,182}$ recently measured in ISOLDE and the literature values was verified as the technique of in-source laser ionization spectroscopy was used for the first time to study the mercury isotopes. From this verification it could be concluded that the measurements performed in 2015 are reliable.

All the other isotopes were analysed by the collaboration. The preliminary results of the changes in mean square charge radii $\delta < r^2 >$ in the isotopic chain $^{177-208}\text{Hg}$, relative to ^{198}Hg , denote that the regular and smooth variation of $\delta < r^2 >$ is restored for the neutron-deficient mercury isotopes behind ^{181}Hg . This indicates that the ground state returns to a slightly deformed oblate state as expected from theoretical considerations which presume that $N = 82$ indicates in this entire region a shell closure. Therefore one can conclude that this observation supports the assumption of $N = 82$ remaining a magic number even if one approaches the proton dripline.

A key element in the laser spectroscopy measurements is the precise determination of the laser wavelength. However, during the past year a number of problems were identified in two wavelength meter models (i.e. WS-7 and ATOS) that are widely used at several laser laboratories; viz. there were problems of consistency between both wavelength meters in the measurement of the high energy singlet peak of ^{215}Ac and the hyperfine spectrum of ^{63}Cu could not be reproduced when using WS-7. As one of the wavelength meters (WS-7) was of the same type as the one used for the mercury laser spectroscopy measurements, a series of systematic measurements to characterize the performance of the wavelength meters were performed in the In-Gas Laser Ionization and Spectroscopy (IGLIS) laser laboratory. The problems could be reduced to: a wrong calibration of the ATOS interferometers which was solved by the manufacturer and a wrong calibration of the WS-7 wavelength meter. The hyperfine spectra of the mercury isotopes were measured with the correct calibration file.

Mean field approaches are trying to describe the odd-mass mercury isotopes. (The description of the odd-mass mercury isotopes is more labour intensive compared to the even-mass mercury isotopes due to difficulties involved in the restoration of the broken symmetries.) Once these calculations are performed, a comparison will be made with the experimental $\delta < r^2 >^{AA'}$ measured in April 2015.

Nucleon transfer reactions can be performed in order to get insight in the underlying microscopic structure of these strongly deformed structures in the odd-Hg isotopes. These experiments can only be performed after the development of intense radioactive mercury beams [2] and the completion of the HIE ISOLDE accelerator in order to reach an energy of 10 MeV/u needed for these few-nucleon transfer reactions.

In June 2016, the knowledge of the mean square charge radius behavior of the bismut isotopes will be extended towards the more neutron-deficient isotopes using the same set-up as the one used for the mercury experiment. These experiments complete the in-source laser ionization spectroscopy measurements in the lead region.

It is interesting to extend the laser spectroscopy experiments towards the heavy and superheavy elements, not only from nuclear physics point of view, but also from atomic physics as little is known about the atomic physics of the heaviest elements. The HELIOS project is developing a new laser spectroscopy method to study the nuclear properties of the heavy and superheavy elements, with as ultimate goal the investigation of nobelium and beyond. The proposed methodology is based on in-gas jet laser ionization spectroscopy that promises to reach the necessary efficiency, selectivity and spectral line width to study this region of the nuclear chart. One believes that shape coexistence occurs in all but the lightest nuclei. Therefore the investigation of this region can eventually help to improve our currently existing knowledge about shape coexistence.

Appendix A

Information about the scans

Information about the scans is reported in this chapter of the appendix. The energy spectra for the silicon detectors are given for each measurement together with the HFS. If suitable gamma lines could be found, the corresponding HFS are given with and without background correction.

A.1 ^{181}Hg

A.1.1 181Hg_scan_0362

The energy spectra for Si1 and Si2 (figure A.1) display some extra alpha lines of the decay of ^{181}Hg compared to the energy spectra of measurement 181Hg_scan_0279_to_0280 (figures 4.1 and A.3). This is explained by the higher amount of observed Hg ions. By integrating over the green areas in figure A.1, the HFS (figure A.2) is obtained.

The procedure explained in section 4.2.1 for determining appropriate gamma lines to make HFS was applied. One X-ray could be used to make a HFS, namely the 42.5 keV line measured by the LEGe. A HFS was made, but this spectra was unclear and not suitable to fit.

There is no information written down in the logbooks of the experiment about this measurement. The data acquisition time was around 40 s. This estimate is based on the amount of pulser pulses of a 100 Hz pulser for the first laser step. This data acquisition time is longer than for measurement 181Hg_scan_0271_to_0274. However there are less counts in the HFS (figure A.2) compared to the HFS for scan 181Hg_scan_0271_to_0274 (figure 4.8). Probably there was a micro beam gate (concept explained in figure 3.11) used. But the delay and the duration of the uBG is unknown. The fewer number of counts compared to measurement 181Hg_scan_0271_to_0274 (which has deadtime) can explain why there was no deadtime for this scan.

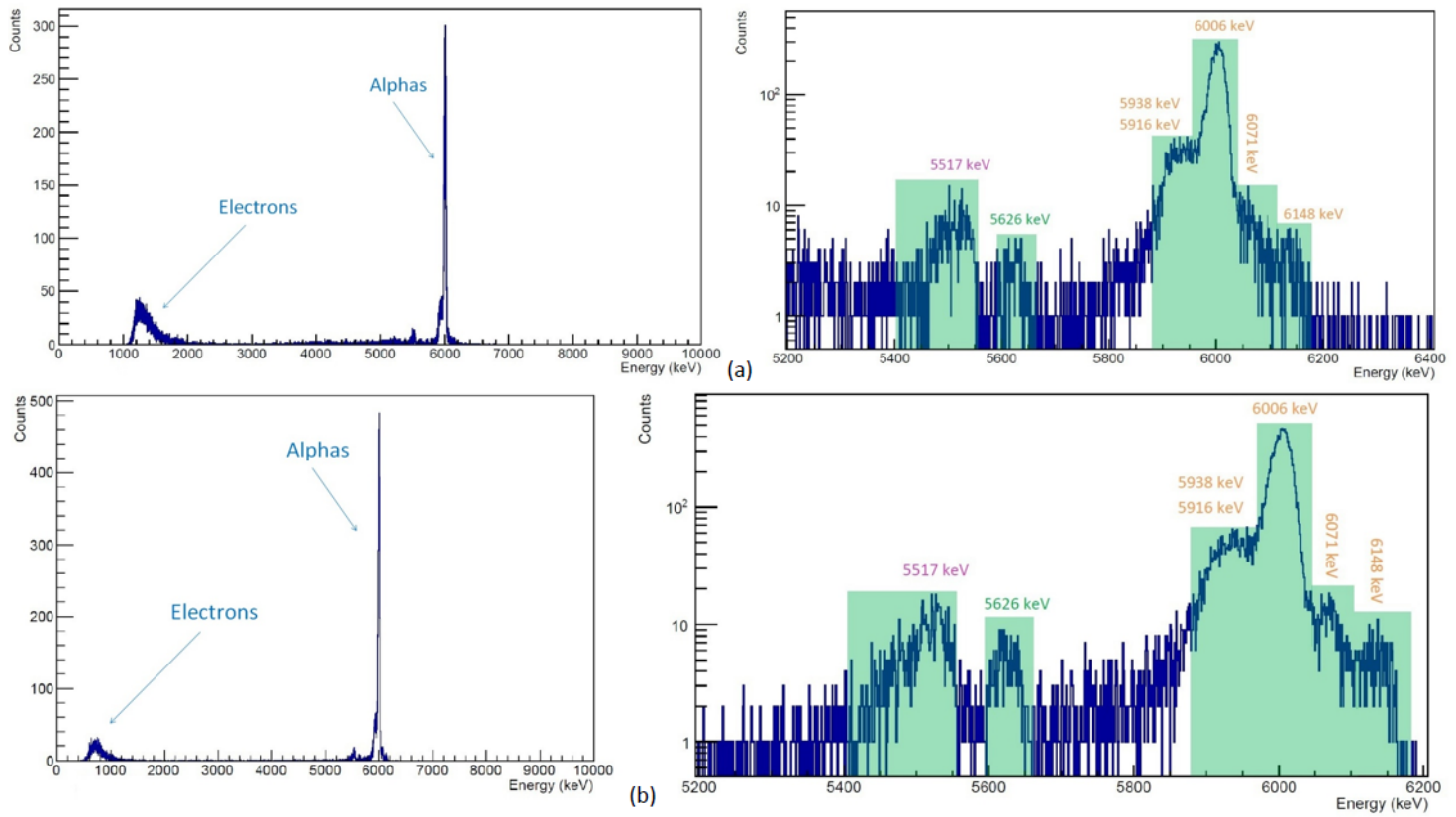


Figure A.1: Energy spectra for (a) Si1 and (b) Si2 of measurement 181Hg_scan_0362 of ^{181}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{181}Hg (figure 1.2a). The green shaded areas represent the areas over which the integration is performed to make the HFS in figure A.2.

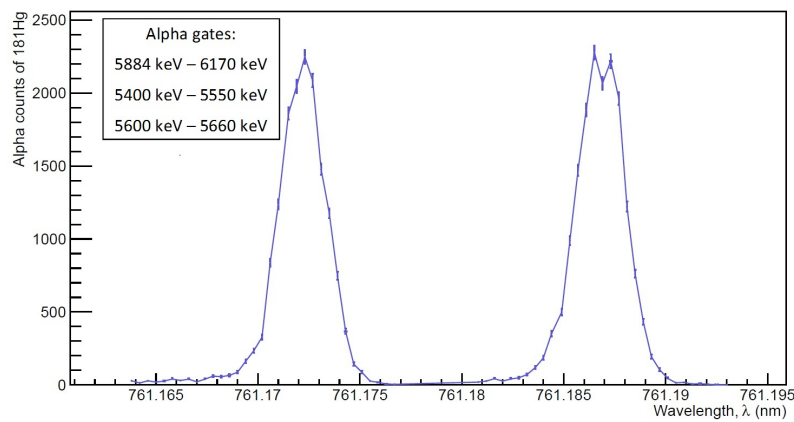


Figure A.2: Alpha HFS for measurement 181Hg_scan_0362 of ^{181}Hg . The used alpha energy gates are displayed in the upper box and are illustrated in figure A.1.

A.1.2 ^{181}Hg scan_0279_to_0280

The energy spectra for Si1 and Si2 are displayed in figures A.3 and 4.1, respectively. The obtained alpha HFS is given in figure 4.2. The procedure explained in section 4.2.1 for determining appropriate gamma lines to make HFS was applied. A HFS obtained by gating on the 42.5 keV line of the β -decay of ^{181}Hg was constructed (figure A.4).

The data acquisition time for this scan was 25100 ms. A micro beam gate (concept explained in figure 3.11) of 100 ms with a delay of 100 ms was used for this scan. Because of this uBG there are less counts in the HFS (figure 4.2) compared to the HFS of scan 181Hg_scan_0271_to_0274 (figure 4.8).

There is no deadtime for this scan. This can be explained by the fact that there are less counts compared to scan 181Hg_scan_0271_to_0274 (which has deadtime). In order to carry out the deadtime correction, it had to be taken into account that the pulser was not connected to the gFLT signal (concept explained in figure 3.10).

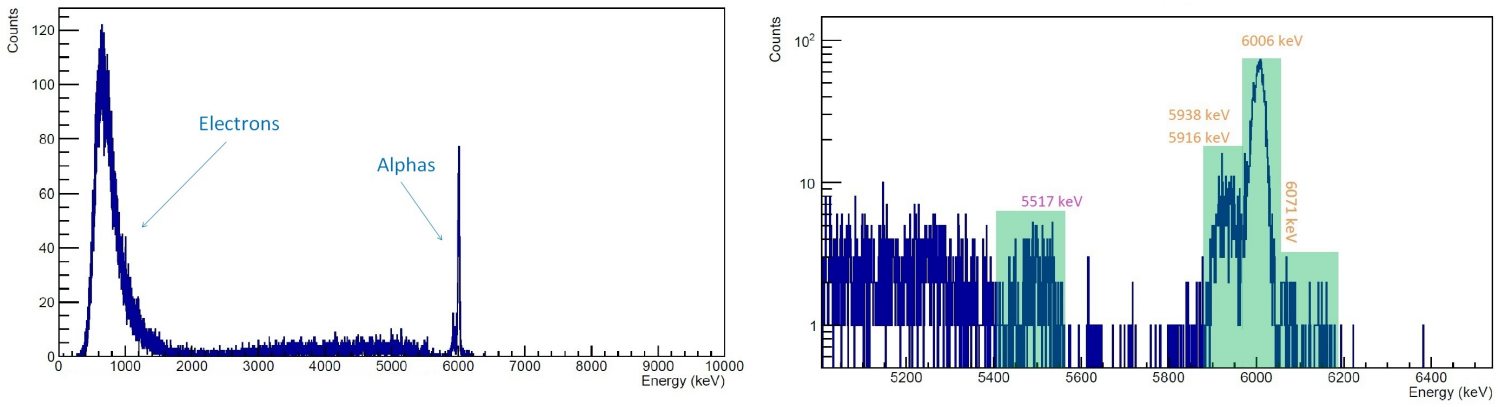


Figure A.3: Energy spectra for Si1 of measurement 181Hg_scan_0279_to_0280 of ^{181}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{181}Hg (figure 1.2a). The green shaded areas represent the areas over which the integration is performed to make the HFS in figure 4.2.

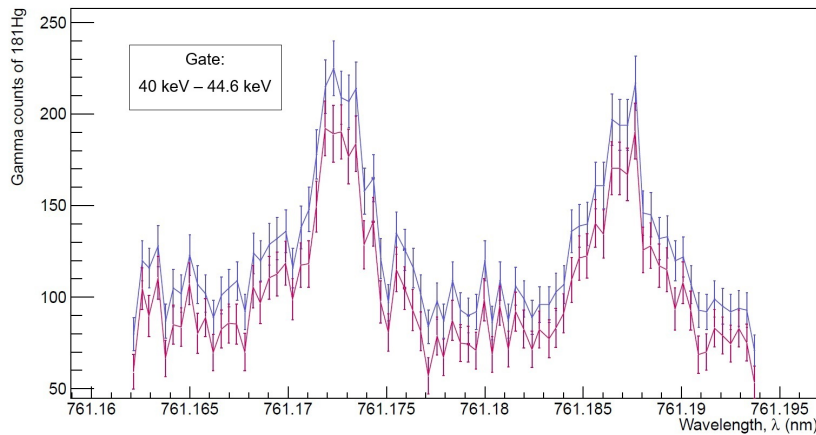


Figure A.4: HFS obtained by gating on the 42.5 keV line from the β -decay of ^{181}Hg taken by LEGe of measurement 181Hg_scan_0279_to_0280, before (blue) and after (pink) background subtraction.

A.1.3 ^{181}Hg scan_0271_to_0274

The energy spectra for Si1 and Si2 (figure A.5) display some extra alpha lines of the decay of ^{181}Hg compared to measurements 181Hg_scan_0279_to_0280 (figures 4.1 and A.3) and 181Hg_scan_0362 (figure A.1). This is explained by the higher amount of observed Hg ions. By integrating over the green areas in figure A.5 the HFS shown in figure 4.8 is obtained. A gamma HFS could be made by gating on the 42.5 keV line of the β -decay of ^{181}Hg (figures 4.8 and A.6).

The data acquisition time for this measurement was 25100 ms.

There is deadtime for this scan. This is explained by the large amount of counts. In order to carry out the deadtime correction, it had to be taken into account that the pulser was not connected to the gFLT signal (concept explained in figure 3.10). The HFS based on counting alpha particles together with the HFS based on counting photons after deadtime correction are shown and explained in section 4.3.

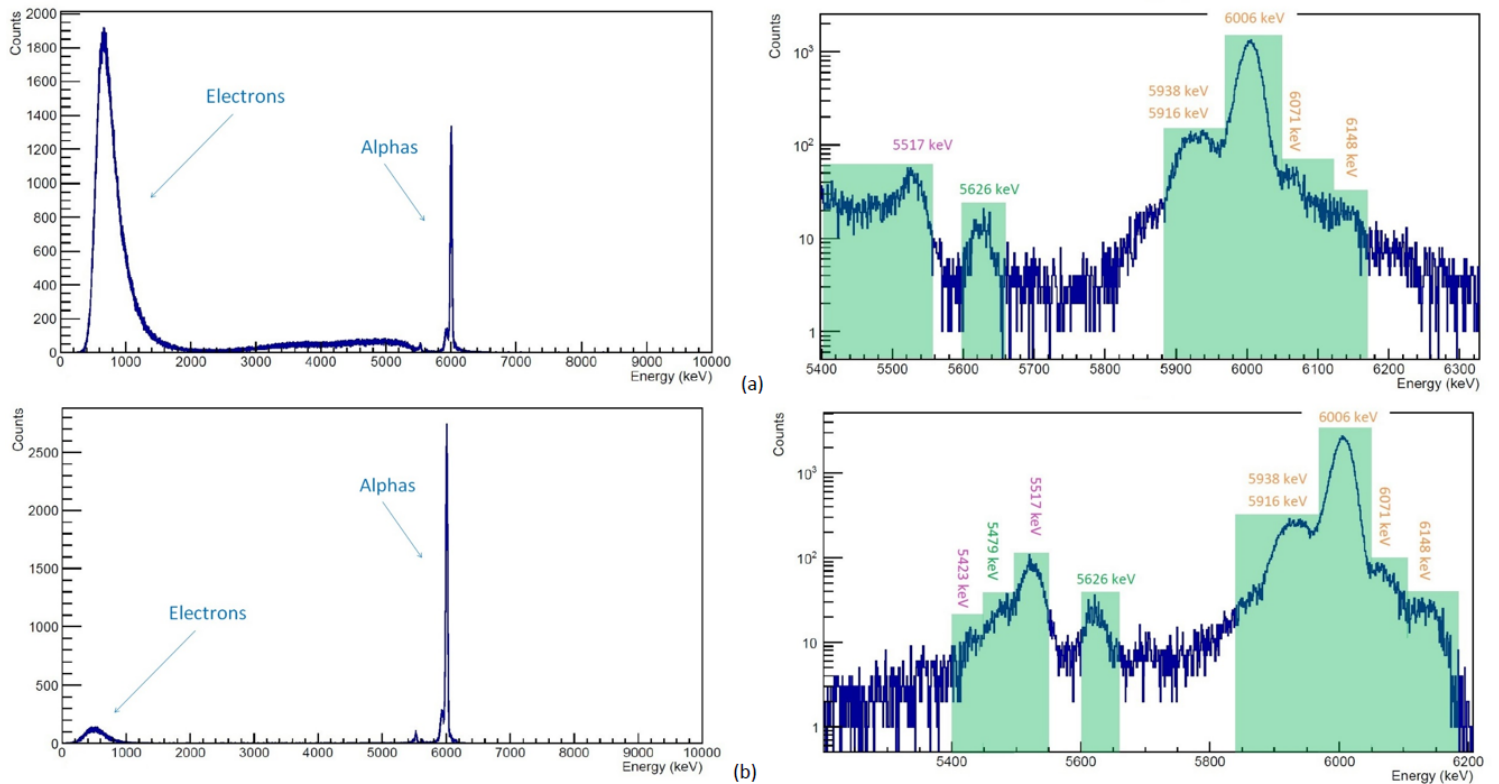


Figure A.5: Energy spectra for (a) Si1 and (b) Si2 of measurement 181Hg_scan_0271_to_0274 of ^{181}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{181}Hg (figure 1.2a). The green shaded areas represent the areas over which the integration is performed to make the HFS in figure 4.8.

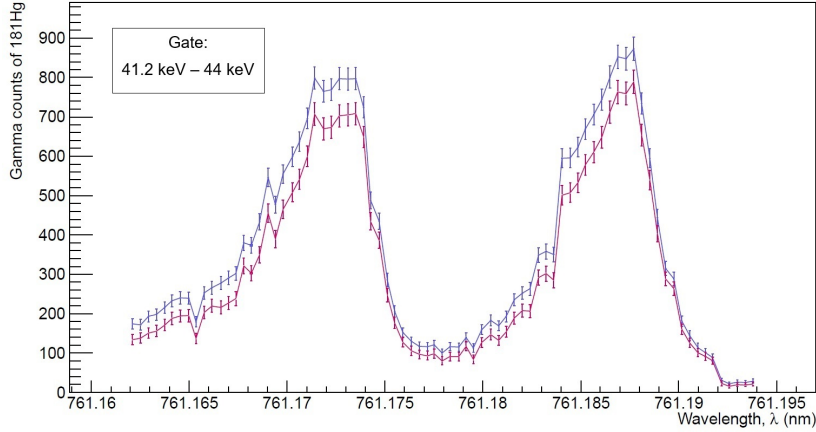


Figure A.6: HFS obtained by gating on the 42.5 keV line from the β -decay of ^{181}Hg taken by LEGe of measurement 181Hg_scan_0271_to_0274, before (blue) and after (pink) background subtraction.

A.2 ^{182}Hg

A.2.1 182Hg_scan_0366

The energy spectra for Si1 and Si2 are shown in figures A.7 and 4.1, respectively. By integrating over the green areas in these figures the HFS (figure 4.2) is obtained.

In total four gamma HFS could be made; using the 129.5 keV line measured with LEGe (figure 4.7), the 217.6 keV line measured with LEGe and Ge2 and the 413.5 keV line measured with Ge2 (together displayed in figure A.8).

The data acquisition time for this scan was 40800 ms. Because the germanium detectors got oversaturated during the previous measurement of ^{182}Hg (scan 182Hg_scan_0364, as explained in section A.2.2) a micro beam gate of 100 ms without delay was used for this scan. The concept of a micro beam gate is explained in figure 3.11. Because of this uBG there are less counts in the HFS (figure 4.2) for this scan compared to the HFS of scan 182Hg_scan_0364 (figure A.10).

There is no deadtime for this scan. This can be explained by the fact that there are less counts compared to scan 181Hg_scan_0271_to_0274 of ^{181}Hg .

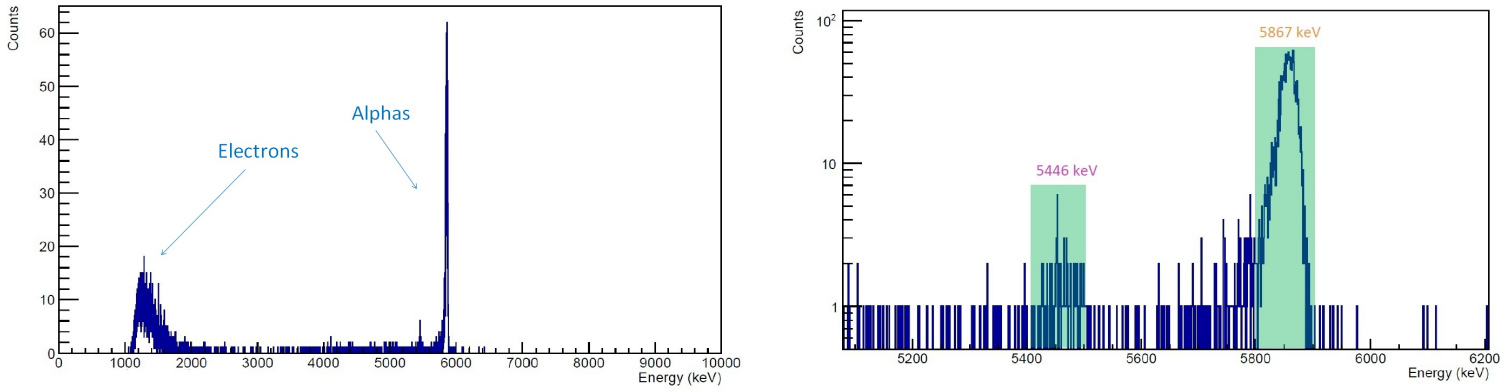


Figure A.7: Energy spectra for Si1 of measurement 182Hg_scan_0366 of ^{182}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{182}Hg (figure 1.2b). The green shaded areas represent the areas over which the integration is performed to make the HFS in figure 4.2.

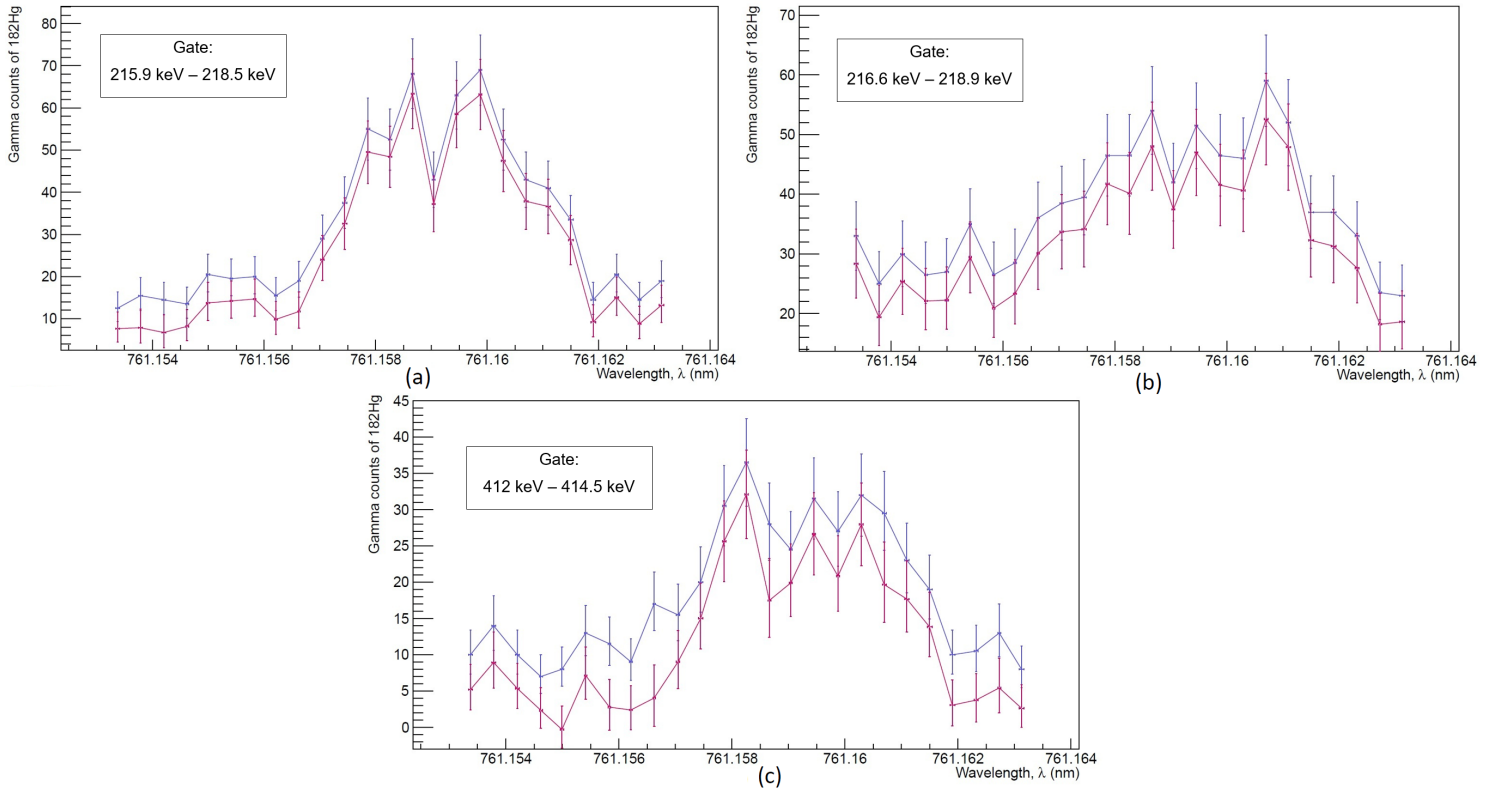


Figure A.8: Gamma HFS for measurement 182Hg_scan_0366 of ^{182}Hg before (blue) and after (pink) background subtraction. The used energy gates are displayed in the upper boxes. (a) 217.6 keV line measured with LEGe. (b) 217.6 keV line measured with Ge2. (c) 413.5 keV line measured with Ge2.

A.2.2 ^{182}Hg scan_0364

The energy spectra for Si1 and Si2 are shown in figure A.9. By integrating over the green areas in this figure the HFS shown in figure A.10 is obtained. A gamma HFS was made for the clearest peak, namely the 217.6 keV line. The result is shown in figure A.11. From this figure it is clear that the germanium detectors got oversaturated during this measurement. Therefore no photon energy gates can be used to make a HFS.

The data acquisition time for this scan was 40800 ms.

It was observed that there was no deadtime correction necessary for this scan. This can be explained by the fact that the germanium detectors got oversaturated.

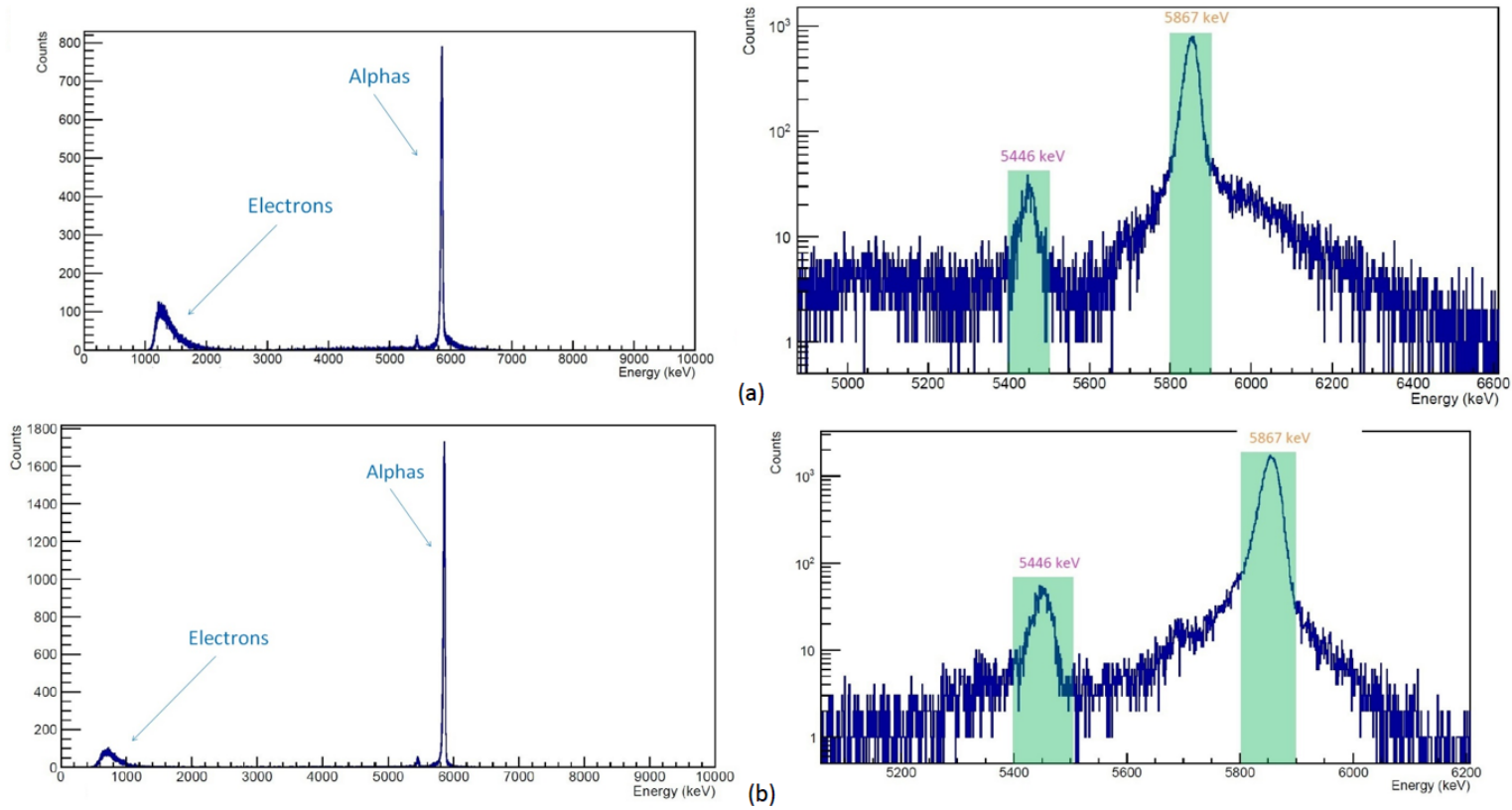


Figure A.9: Energy spectra for (a) Si1 and (b) Si2 of measurement ^{182}Hg _scan_0364 of ^{182}Hg . Left: total energy spectra. Right: energy spectra zoomed in on the alpha particle energy region. The different alpha peaks in the spectrum have been identified and their energy is indicated in a color that corresponds to a specific decay, as explained in the decay scheme of ^{182}Hg (figure 1.2b). The green shaded areas represent the areas over which the integration is performed to make the HFS in figure A.10.

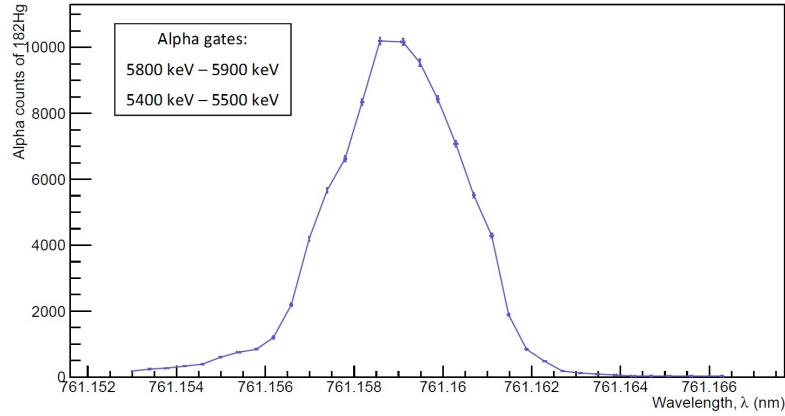


Figure A.10: Alpha HFS for measurement 182Hg_scan_0364 of ^{182}Hg . The used alpha energy gates are displayed in the upper box and are illustrated in figure A.9.

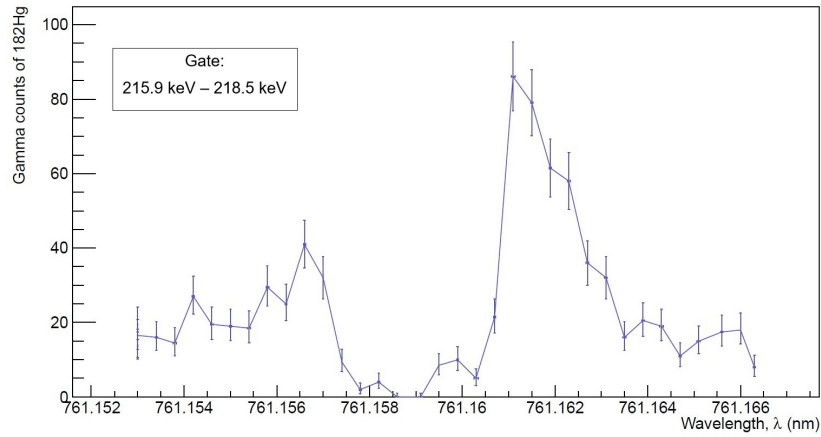


Figure A.11: Gamma HFS for measurement 182Hg_scan_0364 of ^{182}Hg obtained by gating on the 217.6 keV line measured with LEGe. The used energy gate is displayed in the upper box. The almost zero counts at resonance can be explained by the fact that the germanium detectors were overloaded. The germanium detectors received so many photons that they were no longer working properly.

Appendix B

Extra information about the analysis of the HFS

In this chapter of the appendix some extra information and figures belonging to chapter 5 are reported.

B.1 ^{182}Hg

B.1.1 182Hg_scan_0366

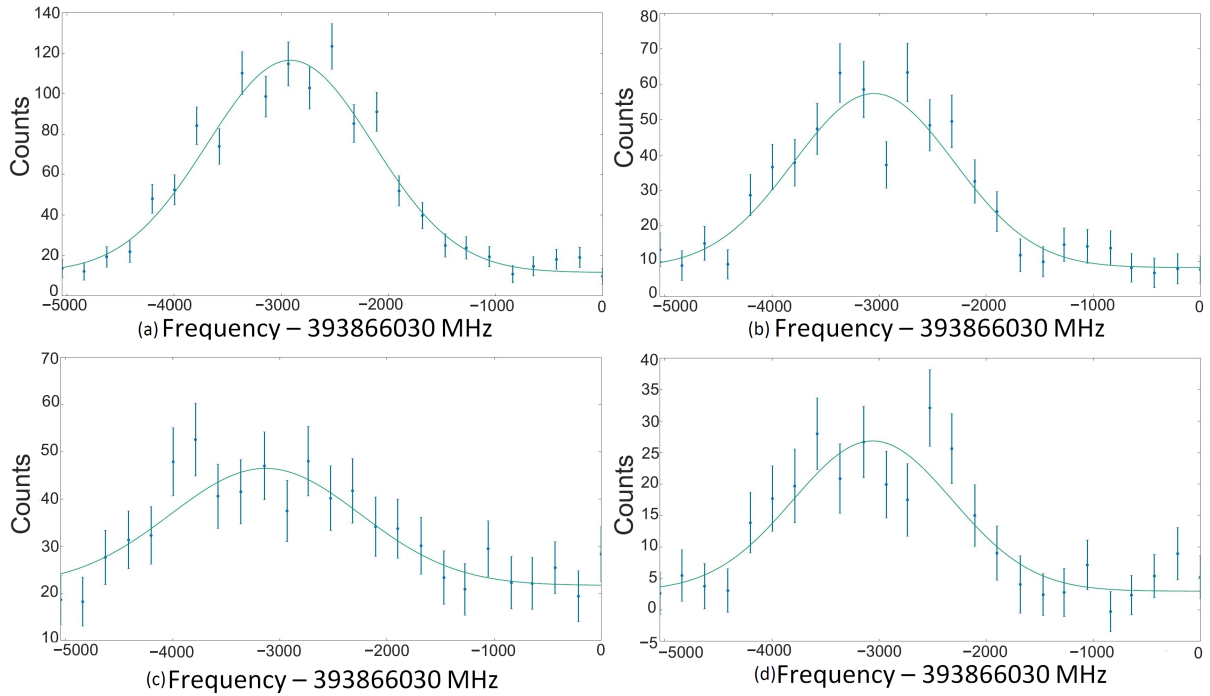


Figure B.1: Fitted gamma HFS for measurement 182Hg_scan_0366 of ^{182}Hg . The frequency difference with the first measured frequency is displayed. (a) HFS obtained by gating on the 129.5 keV line measured with LEGe. Gaussian profile ($\chi^2_{red}=1.2$). (b) HFS obtained by gating on the 217.6 keV line measured with LEGe. Gaussian profile ($\chi^2_{red}=1.2$). (c) HFS obtained by gating on the 217.6 keV line measured with Ge2. Gaussian profile ($\chi^2_{red}=0.7$). (d) HFS obtained by gating on the 413.5 keV line measured with Ge2. Gaussian profile ($\chi^2_{red}=0.9$).

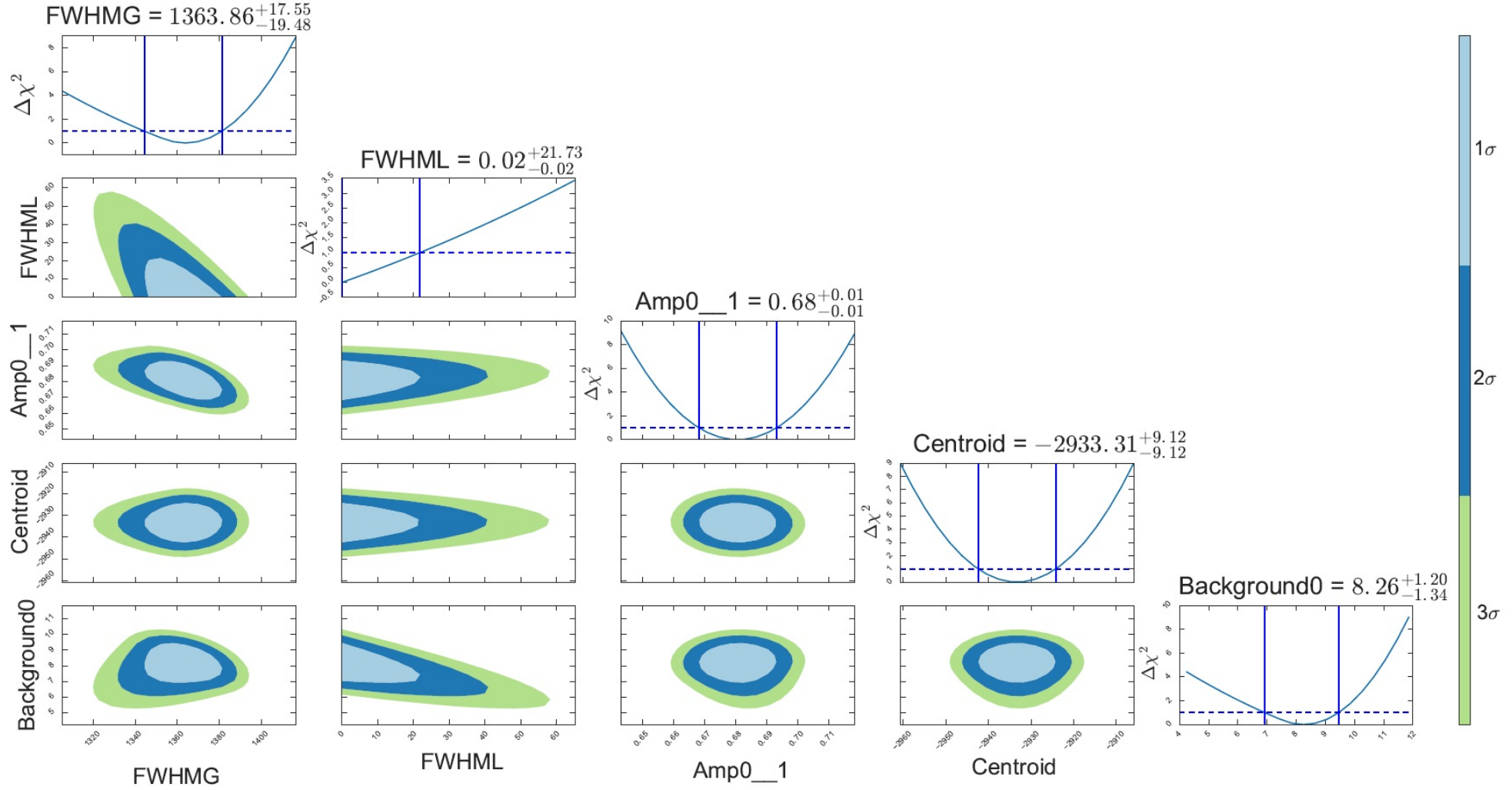


Figure B.2: Correlation plot obtained for measurement 181Hg_scan_0366 of ^{181}Hg by performing a χ^2 -fit with a Voigt profile. The plots on the diagonal are the calculated χ^2 -values for different fixed values of one parameter while refitting the rest of the parameters. In the ideal case, these plots should be parabolas. However this is not the case for the full width at half maximum of the Lorentzian (FWHML) in the Voigt profile. Some problems arise from the fact that the fit wants to let the FWHML, which is restricted to positive values, go to zero. On the off-diagonal contour plots with 1,2 and 3 sigma contours are displayed. From this correlation plot it can be concluded that the Voigt profile is not the suited one to use. Instead a Gaussian profile is used.

B.2 ^{181}Hg

B.2.1 181Hg_scan_0271_to_0274

There is one gamma HFS available for this measurement, which is obtained by gating on the 42.5 keV line of the β -decay of ^{181}Hg measured with the LEGe detector. This HFS has a big tail in both resonance peaks (see figure 4.8).

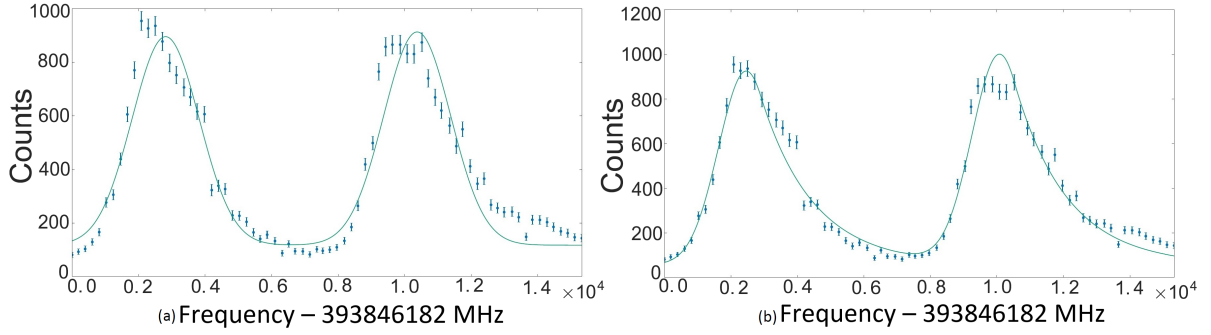


Figure B.3: Fitted gamma HFS obtained by gating on the 42.5 keV line taken by LEGe for measurement 181Hg_scan_0271_to_0274 of ^{181}Hg . The frequency difference with the first measured frequency is displayed. (a) Gaussian profile ($\chi^2_{red} = 14.0$). (b) Crystalball profile ($\chi^2_{red} = 6.5$).

When the HFS is fitted with a Gaussian profile (see figure B.3), the fit function is shifted towards the tails. Therefore it was decided to fit this HFS with a Crystalball profile (see figure B.3). This function consist of a Gaussian with an exponential tail and is mainly used to describe energy loss processes.

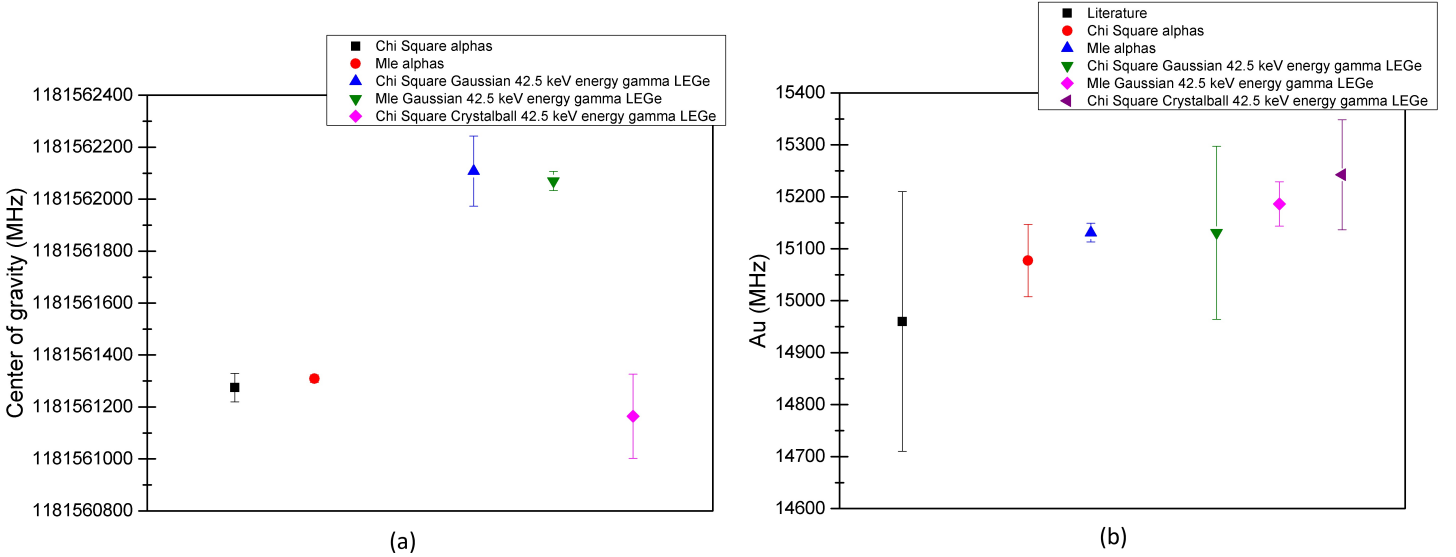


Figure B.4: Results for (a) the center of gravity and (b) the magnetic dipole interaction parameter of the upper level (A_u) of ^{181}Hg , after tripling the fit values, for both χ^2 -fitting (including χ^2_{red} -correction) and mle-fitting derived from the alpha and gamma HFS for measurement 4444 of ^{181}Hg . Literature value is taken from [43].

The results for the center of gravity and A_u for both fit profiles are compared with those from the alpha HFS in figure B.4. The values for the center of gravity are in agreement when a Crystalball profile is used. The use of this profile has not much influence on the value for A_u . This can be understood by the fact that the two resonance peaks are shifted by about the same amount when a Gaussian profile is used.

B.2.2 $^{181}\text{Hg_scan_0279_to_0280}$

The gamma HFS obtained by gating on the 42.5 keV line of the β -decay of ^{181}Hg measured with LEGe is fitted with a Gaussian profile (see figure B.5), since the correlation plot for the Voigt profile was not reliable.

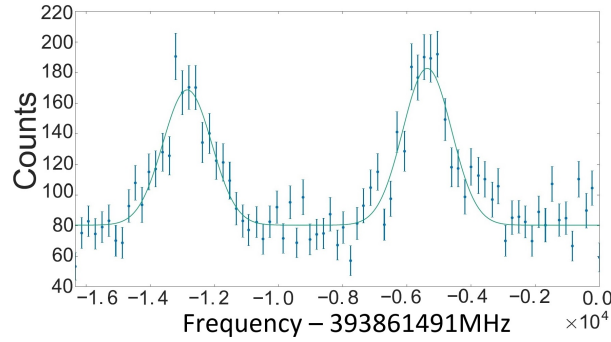


Figure B.5: Fitted gamma HFS obtained by gating on the 42.5 keV line of the β -decay of ^{181}Hg taken by LEGe for measurement 181Hg_scan_0279_to_0280. A Gaussian profile is used ($\chi^2_{red} = 1.6$). The frequency difference with the first measured frequency is displayed.

A comparison between the results for the center of gravity and A_u originating from the gamma and alpha HFS is given in figure B.6. The values for A_u agree well, while the results for the center of gravity agree within two standard deviations.

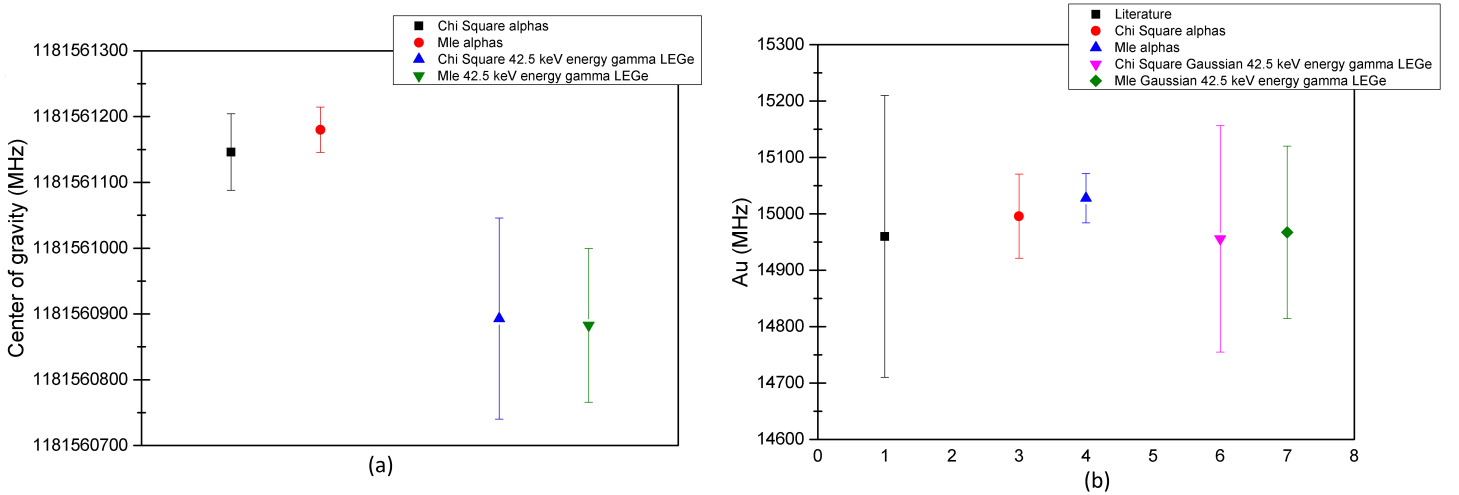


Figure B.6: Results for (a) the center of gravity and (b) the magnetic dipole interaction parameter of the upper level (A_u) of ^{181}Hg , after tripling the fit values, for both χ^2 -fitting (including χ^2_{red} -correction) and mle-fitting derived from the alpha and gamma HFS for measurement 0000 of ^{181}Hg . Literature value is taken from [43].

Appendix C

Windmill measurements at ISOLDE

In at least two articles [37] [38] it is stated that the distance between Si2 and the carbon foil is 3-4 mm. This results in an expected absolute efficiency around 30%, which is the solid angle at this distance. The absolute efficiency for Si detectors equals the solid-angle, because the intrinsic efficiency for the detection of alphas is close to 100%.

The efficiency of Si2 was missing a factor of 2 from what was expected. In November 2015 efficiency measurements on the Si detectors in the Windmill system were performed in order to solve this problem.

The problem was solved by very accurately measuring the distance between Si2 and the Carbon foil. This distance was around 8 mm whereas it was previously thought to be 3-4 mm. This distance is confirmed by the technical drawings of the WM system.

In figure C.1 the different distances that are measured are indicated. Photos of the different components in this drawing are shown in figure C.2. The distance between Si2 and the carbon foil equals 7.8 mm ($= 16.83 \text{ mm} - 2.43 \text{ mm} - 10.1 \text{ mm} + 1.1 \text{ mm} + 2.4 \text{ mm}$) as shown in figure C.1.

After this problem was solved, it was decided to perform efficiency measurements on all the Si detectors in order to make sure that the detectors are working fine. The used detectors are the same as those of the Hg experiment in April 2015, however the positions of Si3 and Si4 were interchanged. The serial number of the detectors is given in table C.1. A ^{241}Am α -calibration source (with serial number NX532) was mounted on the wheel (see figure C.2d) and put in front of the detectors. The main α energy peak at 5486 keV is in coincidence with a 60 keV energy γ -transition to the ground state [40]. In this case, the number of observed γ counts N_γ and the amount of α - γ counts $N_{\alpha\gamma}$ can be used to determine the efficiency of the Si detector ϵ_{Si} as

$$\epsilon_{\text{Si}} = \frac{N_{\alpha\gamma}}{N_\gamma}. \quad (\text{C.1})$$

The obtained efficiencies during this run are summarized in table C.1. These values have to be verified every experiment as the distances in the set-up can vary a few mm and this has a large influence on the efficiencies.

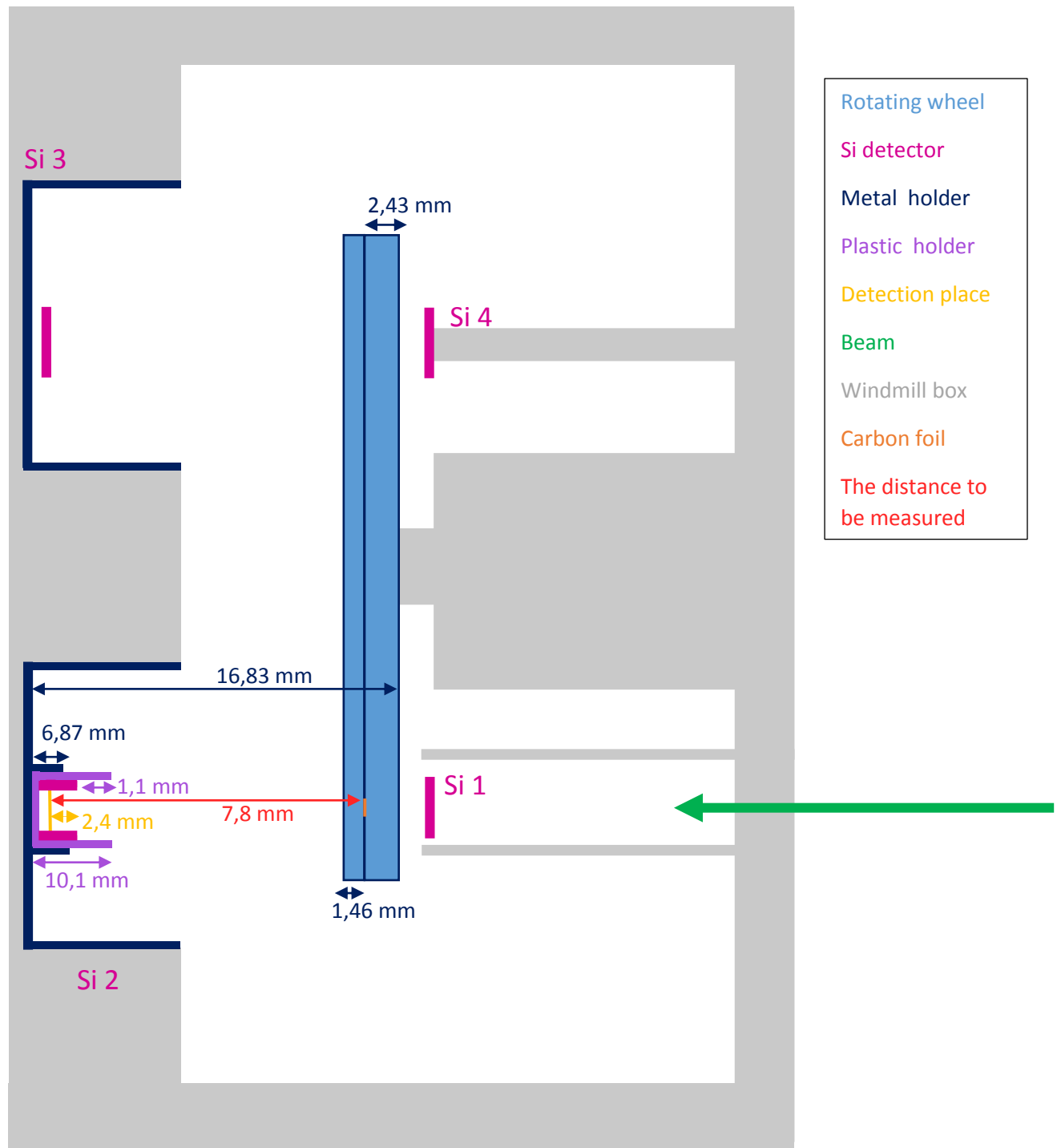


Figure C.1: Cross-section of the Windmill box. This drawing is not on scale. The different distances measured are indicated. Photos of the different components are shown in figure C.2. It is clear from this drawing that the distance between Si2 and the carbon foil was 7.8 mm for the set-up in November 2015.

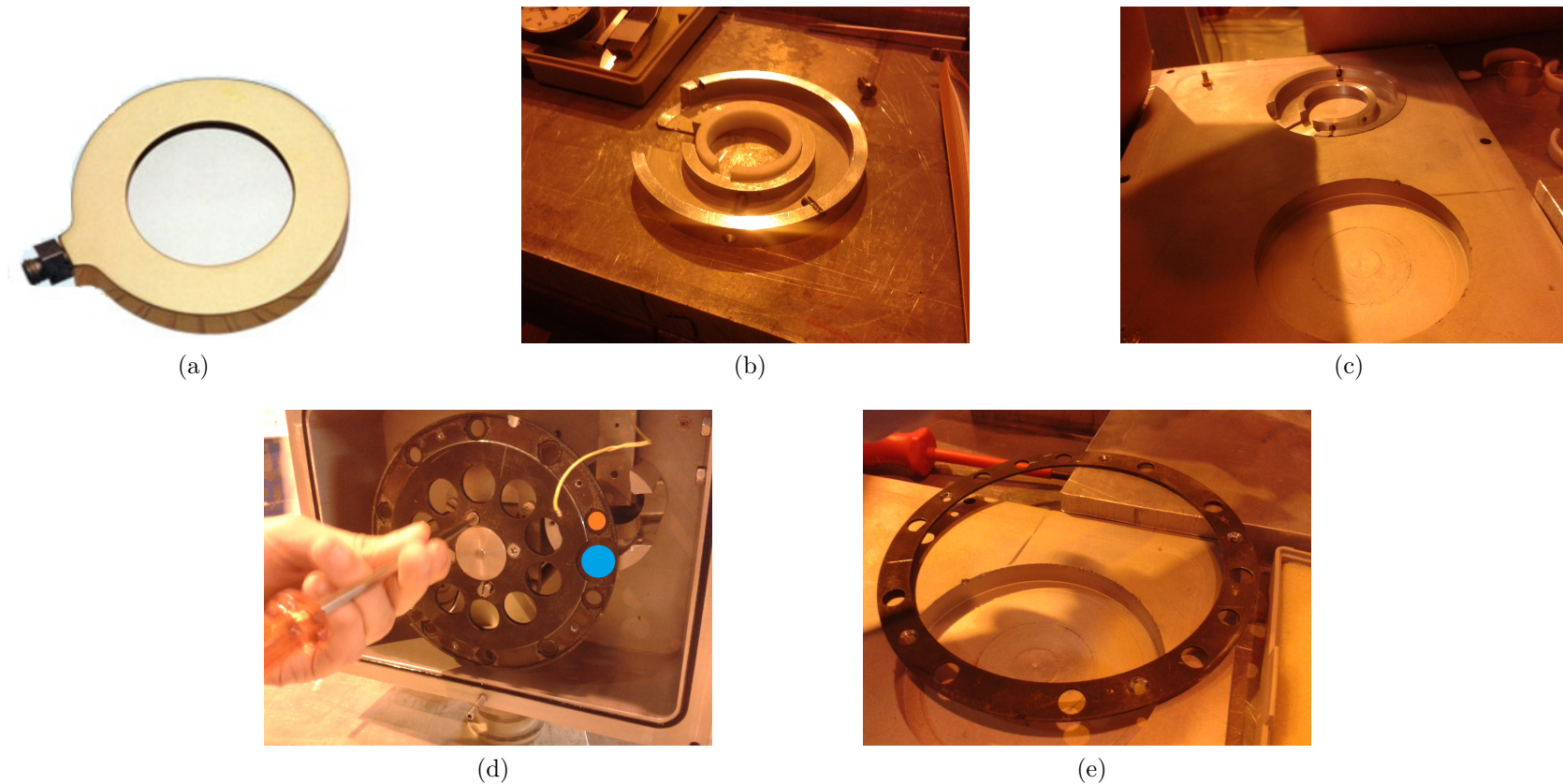


Figure C.2: The different components that have to be measured in order to know the distance between Si2 and the carbon foil inside the Windmill box. (a) Example of a Si detector. The gray area is the sensitive part of the detector. For Si2, it is 2.4 mm below the edge of the detector, as shown in figure C.1. In this figure the detection place is displayed in yellow and the casing in pink. (b) The Si detector is surrounded by a plastic ring before it is placed inside a metal holder. The plastic ring is purple and the metallic holder is dark blue in figure C.1. (c) The metal holder is placed inside one of the holes in the rear plate of the Windmill box. (d) The rotating wheel can be dismantled. When the thing ring, shown in figure C.2e, is removed carbon foils (a possible position is displayed with an orange circle) and two calibration sources (one of the positions is displayed with a blue circle) can be inserted. The position of the carbon foil at the implantation station is displayed in figure C.1 by an orange line. (e) This thing ring fits on the rotating wheel, which is shown in figure C.2d.

Detector	Serial number	Efficiency (%)
Si1	43 082H	13.3
Si2	49 186B	14.6
Si3	74 257	21.2
Si4	74 256	18.7

Table C.1: Obtained efficiencies for the Si detectors in November 2015.

The efficiency of Si1, an annular detector, has a maximum at a certain distance as indicated in figure C.3. In this figure the solid angle is displayed for two annular detectors which are available at KU Leuven, one with a 6 mm hole and another with a 8 mm hole inside. Si1 was not at his optimum distance during the efficiency measurements, but this distance can be used to improve the efficiency during new experiments.

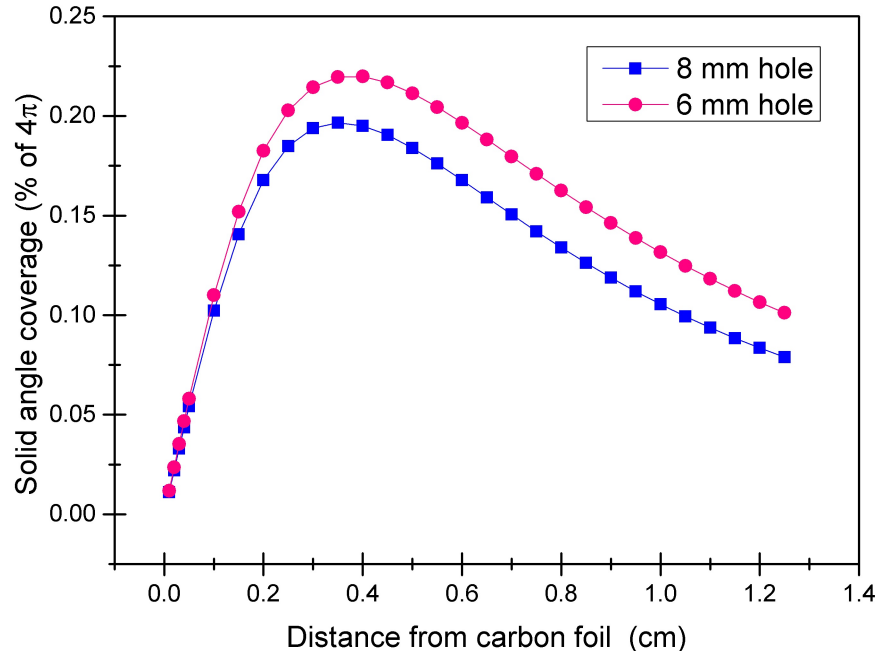


Figure C.3: Annular detector solid angle coverage. Blue: Annular detector with a 6 mm hole. Red: Annular detector with a 8 mm hole.

Appendix D

Determination of the systematic error on the Free Spectral Range of the Scanning Fabry-Pérot Interferometer

The SFPI in the laser laboratory has a FSR of 4002 MHz as stated by the manufacturer. In order to determine the systematic error on this value the readout of the SFPI for two wavelengths of the CW diode laser, which are separated by 8 times 4002 MHz measured by the WS-7 wavelength meter, are compared. If the FSR is really 4002 MHz, the signals of those two wavelengths should coincide. However this is not the case as is illustrated in figure D.1. A small time difference ($\Delta t_{CW1} - \Delta t_{CW2}$) is observed. This time difference can be converted to a difference in energy ($E_{CW1} - E_{CW2}$) by assuming linearity and thus

$$E_{CW1} = \frac{\Delta t_{CW1}}{\Delta t_{HeNe}} * 4002 * \frac{\lambda_{HeNe}}{\lambda_{CW}} \quad (D.1)$$

$$E_{CW2} = \frac{\Delta t_{CW2}}{\Delta t_{HeNe}} * 4002 * \frac{\lambda_{HeNe}}{\lambda_{CW}} \quad (D.2)$$

with λ_{HeNe} the wavelength of the HeNe and λ_{CW} the wavelength of the CW diode laser. This difference in energy determines together with the error on the readout of the WS-7 wavelength meter the systematic error on the FSR ($Error_{FSR}$), namely

$$Error_{FSR} = \sqrt{\left(\frac{E_{CW1} - E_{CW2}}{\text{Number of FSR shifted}} \right)^2 + (Error_{\lambda_{CW1}})^2 + (Error_{\lambda_{CW2}})^2}. \quad (D.3)$$

The number of FSR shifted should be as large as possible, since then the time difference is larger and the fit will be less influenced by the small fluctuations in the time positions as given by the oscilloscope. In this measurement the frequency was shifted by 8 FSR's since this was the largest shift possible with the CW diode laser.

Since a difference between wavelengths as measured by the WS-7 is considered, the systematic error on the wavelength readout cancels out and only the statistical error should be taken into account. This statistical error was determined by measuring the wavelength of the HeNe laser over a long period. The fluctuations observed during a time period equal

to the duration of the measurement, which was around one hour for the measurements in section 6.3, has to be used.

In total three measurements of the HeNe wavelength with WS-7 were performed on three different days. The longest measurement took around 16 hours and the results are shown in figure D.2a. Since these measurement results do not have a Gaussian distribution (see figure D.2b), the statistical error on the readout is determined from the difference between the maximum and minimum value rather than the standard deviation. Thus every measurement was divided into parts of an hour. For every part the difference between the minimum and the maximum readout was calculated and the error used in formula D.3 was set to the maximal difference observed, which was equal to 12 MHz.

With this value formula D.3 gives 18 MHz for the systematic error on the FSR.

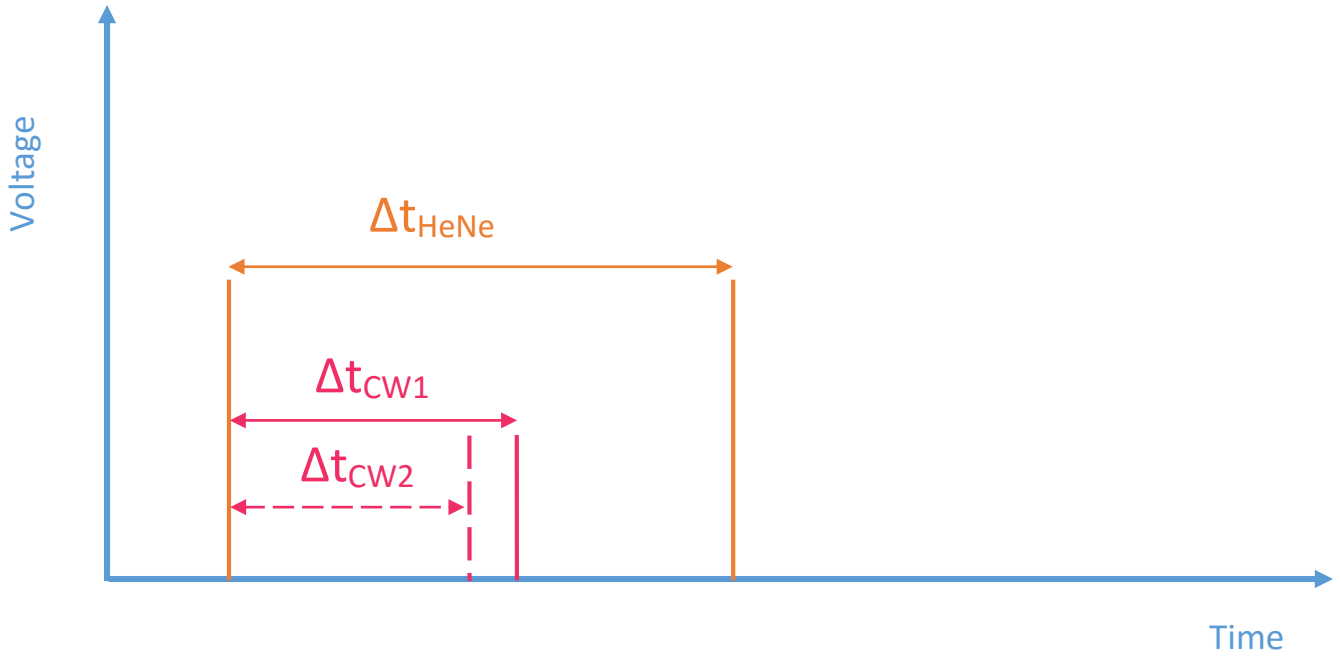


Figure D.1: Illustration of the method of how to determine the systematic error on the FSR of the SFPI. During the measurement the oscilloscope was zoomed to the first order. Orange lines: HeNe signals. Solid line: signal of the first wavelength of the CW diode laser. Dotted line: signal after increasing the frequency by 8 times 4002 MHz. This figure is not to scale.

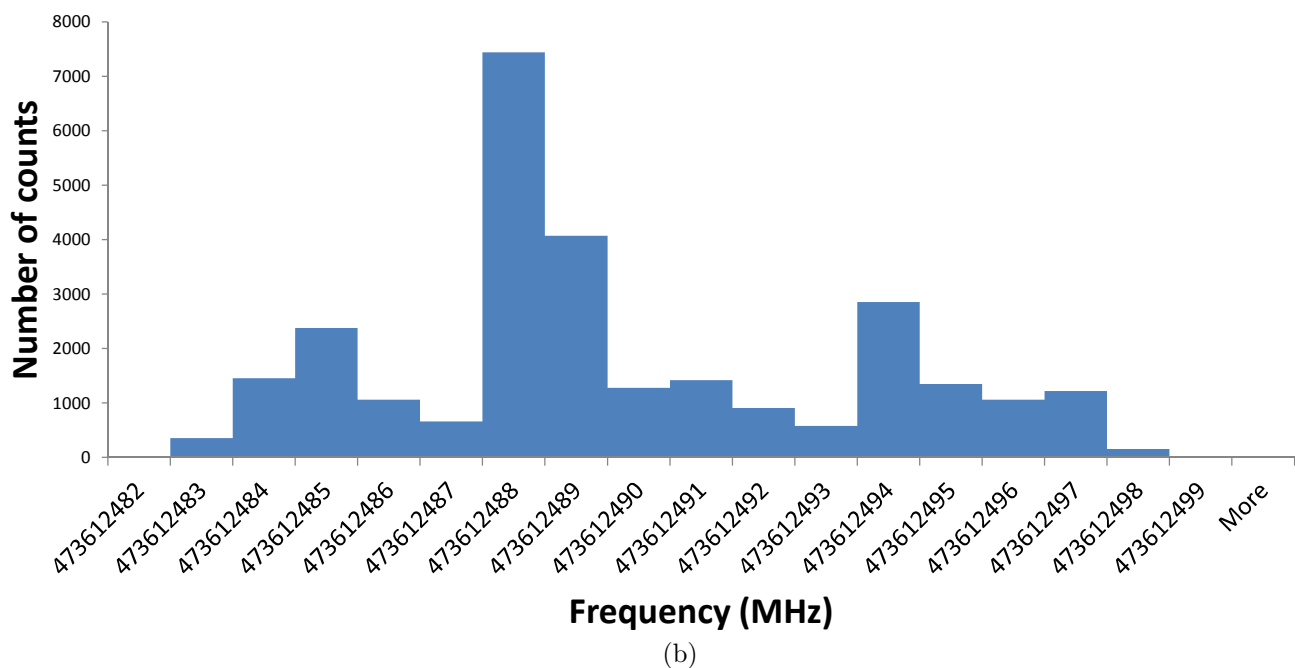
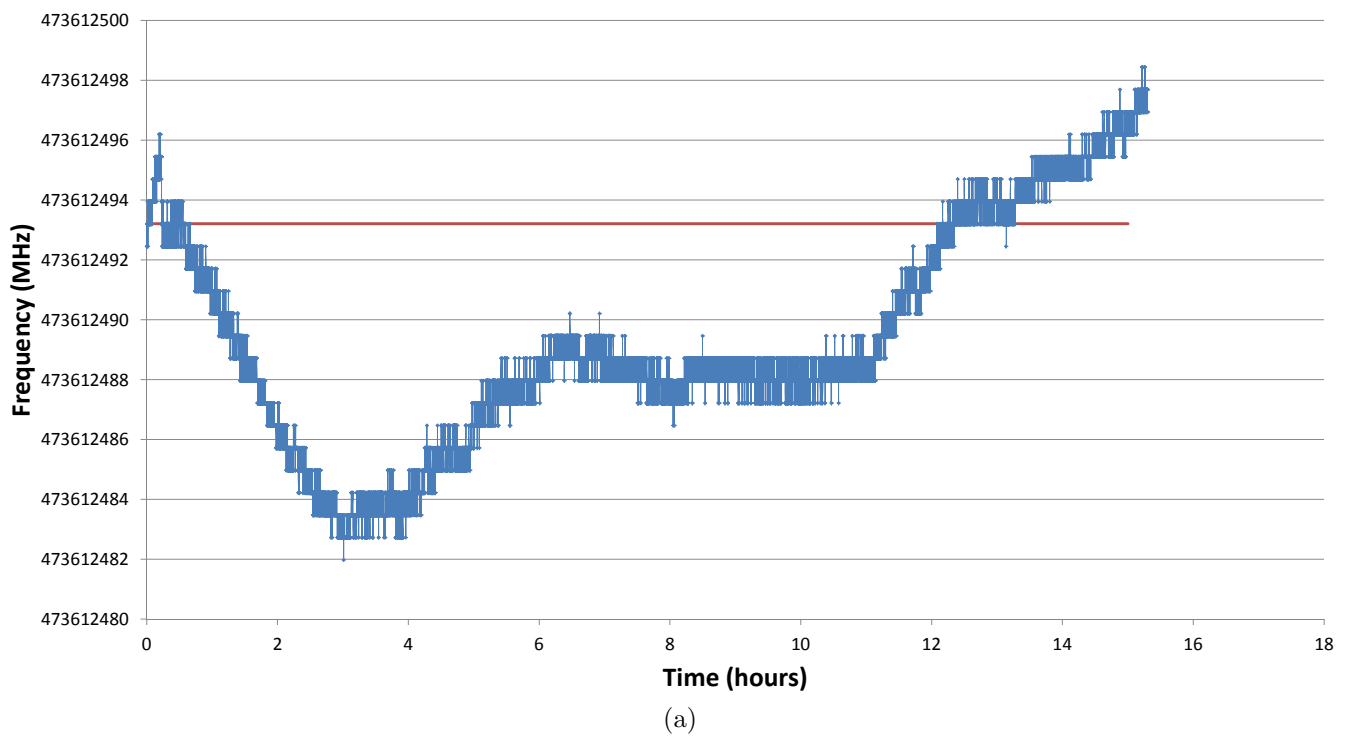


Figure D.2: (a) HeNe wavelength measured by WS-7 during a time period of about 16 hours. The calibration value is indicated by the red line. It is not known what the origin is of the observed drifts, but it can be due to day-night fluctuations. (b) Histogram showing the measured wavelengths of part (a). The distribution is not Gaussian because of the observed drifts.

Bibliography

- [1] N. Kesteloot. Coulomb excitation of ^{200}Po studied at REX – ISOLDE with the Miniball γ spectrometer. Master’s thesis, IKS, KU Leuven, 2010. URL <https://fys.kuleuven.be/iks/ns/files/thesis/nkmasterthesis.pdf>.
- [2] M. Huyse. The Why and How of Radioactive-Beam Research. *Lect. Notes Phys.*, 651:1–32, 2004. doi: 10.1007/978-3-540-44490-9_1. URL http://www.euroschoolonexoticbeams.be/site/files/nlp/LNP651_contrib1.pdf.
- [3] K.S. Krane. *Introductory Nuclear Physics*. John Wiley & Sons, 1988.
- [4] National Nuclear Data Center (nndc) website, 2015. URL <http://www.nndc.bnl.gov/>.
- [5] W.D. Myers and K.-H. Schmidt. An update on the droplet-model charge distribution. *Nuclear Physics A*, 410:61–73, 1983. doi: 10.1016/0375-9474(83)90401-3. URL <http://www.sciencedirect.com/science/article/pii/0375947483904013>.
- [6] D. Berdichevsky and F. Tondeur. Nuclear Core Densities, Isotope Shifts, and the Parametrization of the Droplet Model. *Zeitschrift für Physik A - Atoms and Nuclei*, 322:141–147, 1985. doi: 10.1007/BF01412027. URL <http://link.springer.com/article/10.1007/BF01412027>.
- [7] T. Duguet. Theoretical Nuclear Physics course material, 2015. Unpublished work.
- [8] T. Otsuka. Shell Structure of Exotic Nuclei. *Lect. Notes Phys.*, 764:1–25, 2009. doi: 10.1007/978-3-540-85839-3_1. URL http://www.euroschoolonexoticbeams.be/site/files/nlp/LNP764_contrib1.pdf.
- [9] K. Heyde and J.L. Wood. Shape coexistence in atomic nuclei. *Reviews of Modern Physics*, 83:1467–1521, 2011. doi: 10.1103/RevModPhys.83.1467. URL <http://journals.aps.org/rmp/pdf/10.1103/RevModPhys.83.1467>.
- [10] Web formulas website, 2016. URL http://www.web-formulas.com/Math_Formulas/Geometry_Surface_of_Ellipsoid.aspx.
- [11] J. Wood. Nilsson model course material, 2016. Unpublished work.
- [12] R. B. Firestone. *Table of Isotopes*. Wiley-Interscience, 1996.
- [13] S. Frauendorf and V.V. Pashkevich. On oblate-prolate transition in the ground state rotational band of light mercury isotopes. *Physics Letters B*, 55:365–368, 1975. doi: 10.1016/0370-2693(75)90360-3. URL <http://www.sciencedirect.com/science/article/pii/0370269375903603>.

- [14] G. Ulm et al. Isotope shift of ^{182}Hg and an update of nuclear moments and charge radii in the isotope range ^{181}Hg - ^{206}Hg . *Zeitschrift für Physik A Atomic Nuclei*, 325:247–259, 1986. doi: 10.1007/BF01294605. URL <http://link.springer.com/article/10.1007%2FBF01294605>.
- [15] L. Gaffney. Proposal to the ISOLDE and Neutron Time – of – Flight Committee, In – source laser spectroscopy of mercury isotopes, 2013. Proposal.
- [16] A.E. Barzakh et al. Changes in the mean-square charge radii and magnetic moments of neutron-deficient Tl isotopes. *Physical Review C*, 88:024315, 2013. doi: 10.1103/PhysRevC.88.024315. URL <http://journals.aps.org/prc/pdf/10.1103/PhysRevC.88.024315>.
- [17] A.N. Andreyev. A triplet of differently shaped spin zero states in the atomic nucleus ^{186}Pb . *Nature*, 405:430–433, 2000. doi: 10.1038/35013012. URL <http://www.nature.com/nature/journal/v405/n6785/abs/405430a0.html>.
- [18] J.E. Garcia-Ramos and K. Heyde. Nuclear shape coexistence: A study of the even-even Hg isotopes using the interacting boson model with configuration mixing. *Physical Review C*, 89:014306, 2014. doi: 10.1103/PhysRevC.89.014306. URL <http://inwpent5.ugent.be/papers/PhysRevC.89.014306.pdf>.
- [19] J. Bonn et al. Sudden change in the nuclear charge distribution of very light mercury isotopes. *Physics Letters B*, 38:308–311, 1972. doi: 10.1016/0370-2693(72)90253-5. URL <http://www.sciencedirect.com/science/article/pii/0370269372902535>.
- [20] W.J. Tomlinson and H.H. Stroke. NUCLEAR MOMENTS AND ISOTOPE AND ISOMER SHIFTS OF NEUTRON-DEFICIENT MERCURY ISOTOPES 195, 195^m , 194, 193, 193^m and 192. *Nuclear Physics*, 60:614–633, 1963. doi: 10.1016/0029-5582(64)90098-7. URL <http://www.sciencedirect.com/science/article/pii/0029558264900987>.
- [21] S.P. Davis, T. Aung, and H. Kleinman. Isotope Shifts of Neutron-Deficient Mercury Isotopes. *Physical Review*, 147:861–868, 1966. doi: 10.1103/PhysRev.147.861. URL <http://link.aps.org/doi/10.1103/PhysRev.147.861>.
- [22] S. Cottenier and M. Rots. *Hyperfine Interactions and their Applications in Nuclear Condensed Matter Physics: a microscopic introduction*. Springer, 2005.
- [23] W.R. Johnson. Many-Body Calculations of the Isotope Shift. 2001. URL http://www3.nd.edu/~johnson/Publications/i_shift.pdf. Unpublished work.
- [24] R. Neugart and G. Neyens. Nuclear moments. *Lect. Notes Phys.*, 700:135–189, 2006. doi: 10.1007/3-540-33787-3_4. URL http://euroschoolonexoticbeams.be/site/files/nlp/LNP700_contrib4.pdf.
- [25] P. Van Duppen. Exotic Nuclei: Properties and Interactions - Part A course material, 2015. Unpublished work.

- [26] T. K hl et al. Nuclear Shape Staggering in Very Neutron-Deficient Hg Isotopes Detected by Laser Spectroscopy. *Physical Review Letters*, 39:180–183, 1977. doi: 10.1103/PhysRevLett.39.180. URL <http://link.aps.org/doi/10.1103/PhysRevLett.39.180>.
- [27] E.C. Seltzer. K X-Ray Isotope Shifts. *Physical Review*, 188:1916–1918, 1969. doi: <http://dx.doi.org/10.1103/PhysRev.188.1916>. URL <http://journals.aps.org/pr/pdf/10.1103/PhysRev.188.1916>.
- [28] G. Fricke and K. Heilig. *Nuclear Charge Radii*. Springer, 2004.
- [29] T.E. Cocolios. *Single-particle and collective properties around closed shells probed by in-source laser spectroscopy*. PhD thesis, IKS, KU Leuven, 2010. URL <http://fys.kuleuven.be/iks/ns/phd-master-theses>.
- [30] P. Van Duppen. Isotope Separation On Line and Post Acceleration. *Lect. Notes Phys.*, 700:37–77, 2006. doi: 10.1007/3-540-33787-3_2. URL http://link.springer.com/chapter/10.1007/3-540-33787-3_2.
- [31] ISOLDE website, 2015. URL <http://isolde.web.cern.ch/isolde-logos-layouts-and-templates>.
- [32] CERN website, 2015. URL <http://timeline.web.cern.ch/the-laser-ion-source-rilis-is-developed>.
- [33] S. Sels. β -delayed fission and α -decay spectroscopy of $^{194,196}\text{At}$. Master’s thesis, IKS, KU Leuven, 2013. URL <https://fys.kuleuven.be/iks/ns/files/thesis/masterproef-simonsels.pdf>.
- [34] R.N. Wolf et al. ISOLTRAP’s multi-reflection time-of-flight mass separator/spectrometer. *International Journal of Mass Spectrometry*, 349-350:123–133, 2013. doi: 10.1016/j.ijms.2013.03.020. URL <http://www.sciencedirect.com/science/article/pii/S1387380613001115>.
- [35] Ortec website, 2015. URL <http://www.ortec-online.com>.
- [36] Canberra website, 2015. URL <http://www.canberra.com>.
- [37] A.N. Andreyev et al. New Type of Asymmetric Fission in Proton-Rich Nuclei. *Physical Review C*, 105:252502, 2010. doi: 10.1103/PhysRevLett.105.252502. URL <http://journals.aps.org/prl/pdf/10.1103/PhysRevLett.105.252502>.
- [38] J. Elseviers et al. β -delayed fission of ^{180}Tl . *Physical Review C*, 88:044321, 2013. doi: 10.1103/PhysRevC.88.044321. URL <http://journals.aps.org/prc/pdf/10.1103/PhysRevC.88.044321>.
- [39] Wikipedia website, 2016. URL https://en.wikipedia.org/wiki/Gamma_spectroscopy.
- [40] L. Ghys. *β -delayed fission in proton-rich nuclei in the lead region*. PhD thesis, IKS, KU Leuven, 2015. URL <https://fys.kuleuven.be/iks/ns/files/thesis/thesisprintfinal-lars.pdf>.

- [41] SATLAS website, 2016. URL <https://github.com/woutergins/satlas>.
- [42] I.J. Myung. Tutorial on maximum likelihood estimation. *Journal of Mathematical Psychology*, 47:90–100, 2003. doi: 10.1016/S0022-2496(02)00028-7. URL <http://www.sciencedirect.com/science/article/pii/S0022249602000287>.
- [43] J. Bonn et al. Spins, moments and charge radii in the isotopic series ^{181}Hg - ^{191}Hg . *Zeitschrift für Physik A Atoms and Nuclei*, 276:203–217, 1976. doi: 10.1007/BF01412098. URL <http://link.springer.com/article/10.1007/BF01412098>.
- [44] Internal communication with professor Anatoly Barzakh.
- [45] S. Raeder et al. Developments towards in-gas-jet laser spectroscopy studies of actinium isotopes at LISOL. *Nuclear Instruments and Methods in Physics Research B*, 376:382–387, 2016. doi: doi:10.1016/j.nimb.2015.12.014. URL <http://www.sciencedirect.com/science/article/pii/S0168583X15012586>.
- [46] KU Leuven website, 2016. URL <https://fys.kuleuven.be/iks/ns/helios>.
- [47] G. Vinck. Characterization of an atomic beam unit. Master’s thesis, IKS, KU Leuven, 2014.
- [48] Thorlabs website, 2016. URL http://www.thorlabs.de/newgrouppage9.cfm?objectgroup_id=859.
- [49] C. Granados. In gas-cell and in gas-jet laser ionization spectroscopy of actinium isotopes. To be published.
- [50] C. Granados, Ph. D. dissertation, IKS, KU Leuven, In progress.
- [51] Bergström et al. Two different types of hollow-cathode discharges used for high resolution laser spectroscopy on copper. *Zeitschrift für Physik D - Atoms, Molecules and Clusters*, 13:203–206, 1989. doi: 10.1007/BF01436957. URL <http://link.springer.com/article/10.1007%2FBF01436957>.

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