

**Isotope shift and hyperfine structure measurements
on silver, actinium and astatine
by in-source resonant ionization laser spectroscopy**

by

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Abstract

Resonant ionization laser ion sources are applied worldwide to increase purity and intensity of rare isotopes at radioactive ion beam facilities. Especially for heavy elements the laser wavelengths required for efficient resonant laser ionization are not only element dependent, but also vary to small degrees from isotope to isotope. Since the first operation of an actinide target at ISAC-TRIUMF in 2008, the demand for neutron-rich isotopes far away from stability has steadily increased. Those isotopes often have very low production rates so that often only a few ions per second are released. In order to study isotope shifts and hyperfine structure of silver, actinium and astatine, in-source resonant ionization spectroscopy in combination with radioactive decay detection has been applied. Despite the Doppler limited resolution it has the advantage that it is ultra sensitive and the atomic spectrum for the nuclear ground and isomeric states can be investigated individually. An isobaric separation has been demonstrated for $^{115-119}\text{Ag}$, where the hyperfine structure of one state showed a splitting of 22 GHz to 38 GHz while for the other state only a single peak spectrum can be resolved. For astatine and actinium the main interest is to measure and study the optical isotope shift, which is for the first excitation step for neutron-rich isotopes in the order of $\text{IS}_{\text{FES}} \approx \pm 3.7 \text{ GHz u}^{-1}$ for both elements, as these observables give insight into nuclear moments and shape. In addition, also the isotope shift of the second excitation step for astatine has been measured to $\text{IS}_{\text{SES,At}} \approx -1.7 \text{ GHz u}^{-1}$. Laser spectroscopy on astatine has mainly been performed on the neutron-deficient isotopes $^{199,205}\text{At}$ due to high count rates and low isobaric contamination. With the results obtained it is possible to extrapolate the required wavelength for ionizing and delivering the isotopes $^{221-225}\text{At}$ which are of interest to e.g. electric dipole moment studies.

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1 | Introduction

Just over a hundred years ago, it was thought that a hydrogen atom consisted of 1837 electrons in some kind of positively-charged goo [1]. With the discovery of the atomic nucleus by Geiger, Marsden and Rutherford in 1909 [2], the hydrogen atom is now known to actually consist of a single electron orbiting one positively charged proton comprising the main contribution to the atomic mass. Presently, properties of the atomic nuclei find applications in many different areas of daily life; e.g., power generation in nuclear reactors, diagnosis and treatment of certain diseases, or determining the age of organic material via radiocarbon dating. Even though nuclear physics is an integral part of our lives, the properties of the atomic nucleus are not yet fully understood.

Of the more than 3000 nuclei discovered so far only 288 are stable or have half-lives longer than the age of the earth. Based on this fact, it is understandable that especially those nuclei not found on earth or only in very small amounts are of interest to get a deeper understanding of their properties. Such rare isotopes are provided by various radioactive ion beam (RIB) facilities all over the world. Radioactive isotopes can be produced either in a nuclear reactor or by bombarding a target with accelerated ions. For RIB facilities, particle accelerators are commonly used, where we distinguish between two different methods: in-flight versus Isotope Separator On-Line (ISOL) techniques; each having its own advantages and disadvantages. In both cases, the isotope of interest is separated from the variety of simultaneously produced isotopes and transported away from the production area to the experiment.

The in-flight method uses accelerated heavy ions impinging on a thin target. The projectile can undergo fission, fusion or fragmentation reactions during the interaction with the target. The resulting RIB consists of a variety of isotopes. In order to select the isotope of interest, the RIB passes through a fragment separator. Since the energy of the RIB is already high, this production method is especially of interest for nuclear reaction experiments since no post-acceleration is necessary. Furthermore, this technique provides the largest variety of isotopes since the process is chemistry and half-life independent [3]. Institutes that use the in-flight method include GANIL in France, GSI in Germany, NSCL in the USA, and RIKEN in Japan.

For the ISOL technique, a thick target is bombarded with accelerated light particles such as protons, deuterons or alpha particles. The reaction products are created by spallation, fragmentation or fission and stopped inside the target material. The target container is heated up to 2000 °C in order to allow the neutral radioactive atoms to diffuse out of the target and effuse towards an ion source. Singly-charged ions are then extracted from the target and transported to a magnetic mass separator where the isotope of interest can be selected [4]. This production method is ideal for decay or trapping experiments, where a low beam energy is required. In order to study nuclear reactions, it is possible to post accelerate the RIB in an additional particle accelerator. Compared to the in-flight method, the production yields reached with the ISOL technique can often be higher due to the thicker target and higher primary beam intensities. In addition, the beam quality is better, both in emittance and selectivity. On the other hand, it is not possible to extract isotopes from the target with half-lives shorter than 4 ms, and some elements are not released at all including those which are chemically active or refractory. Examples where the ISOL method is used include ISOLDE-CERN in Switzerland, GANIL and IPN in France, and TRIUMF in Canada, where the research for this work was conducted. A more detailed description of the RIB production and delivery at TRIUMF's Isotope Separator and ACcelerator (ISAC) facility is given in Chapter 2.

For the success of many experiments, intensity and purity of the RIB are essential. At ISOL facilities, these beam properties can be ensured with the right choice of target ion source combinations. In

particular, resonant ionization laser ion sources play an important role in realizing this task as they are unrivaled with respect to selectivity. These ion sources rely on the stepwise resonant excitation of a valence electron above the ionization threshold. A description of resonant photo-excitation and ionization is given in Chapter 3. Since every element has its unique spectroscopic fingerprint, this process is completely element-selective. Using multi-step resonant photo-ionization as an efficient ion source at ISOL facilities was first proposed in 1985 [5] and realized in 1991 at the IRIS laboratory at PNPI, Gatchina, Russia [6]. By now, resonant ionization laser ion sources are operational worldwide at seven different RIB facilities [7]. TRIUMF's resonant ionization laser ion source (TRILIS) is described in Chapter 4.

The efficiency of resonant ionization laser ion sources depends strongly on the chosen excitation schemes. Even though many atomic transitions can be found in online databases [8, 9], the information for higher excited states is particularly incomplete. For that reason, the most efficient excitation scheme for the available laser system should be determined off-line on stable isotopes prior to the on-line implementation.

With high resolution spectroscopy techniques such as laser spectroscopy on fast isotope beams in the order of ≈ 50 keV or trapped atoms [10], it can be seen that the atomic spectrum is not only element-dependent but also differs slightly from isotope to isotope. Changes of the nuclear mass m_A and mean squared charge radius $\langle r^2 \rangle$ lead to a shift of the spectrum. The interaction of the valence electron with the nuclear spin I , and electromagnetic multipole moments, results in a small splitting known as hyperfine structure (HFS). More details about isotope shift (IS) and HFS can be found in Chapter 3. These effects are typically too small to be noticed in resonant laser ion source operation that suffers several GHz of Doppler broadening as a result of the 2000 °C hot ionization region. For heavy elements within the lead-region, however, the optical isotope shift and hyperfine structure splitting can be large enough to be detectable with resonant ionization spectroscopy techniques. The frequencies required for efficient ionization of isotopes far away from stability can differ by several laser linewidths from the known wavelengths, since excitations

schemes are commonly tested on stable isotopes. In most cases, experiments performed at RIB facilities are interested in isotopes far away from stability where the production rates can be as low as a few atoms per second. In order to ensure efficient ionization of those isotopes with low production rates, it is not only important to find a suitable ionization scheme but also to study the isotope shift and hyperfine structure splitting of the applied transitions. For some elements where the HFS is particularly large, it is even possible to separate nuclear ground states from isomeric states adding a further purification factor to the RIB production. This isomeric separation has been demonstrated for select silver isotopes (Chapter 5).

While ISs and HFS splittings might complicate the ionization of rare isotopes, studying these effects enables the extraction of changes in nuclear shapes, sizes, and dipole and quadrupole moments. The changes of the mean squared charge radii for elements around the lead isotope chain shown in Figure 1.1 were extracted from optical isotope shift measurements. Several interesting features can be seen such as a kink at the neutron shell closure $N = 126$, a pronounced odd-even staggering for the neutron-deficient mercury isotopes [12], and an early onset of deformation for the neutron-deficient polonium isotopes [13]. Results for two elements, astatine and actinium, where optical isotope shift data had been missing will be presented in Chapters 6 and 7 of this work. A greater part of the data shown in Figure 1.1 was measured using in-source resonant laser ionization spectroscopy, which despite being Doppler limited is one of the most sensitive laser spectroscopy techniques. This sensitivity results from the high efficiency of counting ions rather than photons as it is used in high resolution spectroscopy methods. In addition, it is also possible to monitor the radioactive decay of the mass-separated RIB as a function of the laser frequency, which allows to measure spectra of isotopes with high isobaric background or study the spectra resulting from the nuclear ground state and isomeric state individually. This spectroscopy method has been applied to the measurements of silver and astatine presented in Chapters 5 and 7 of this thesis.

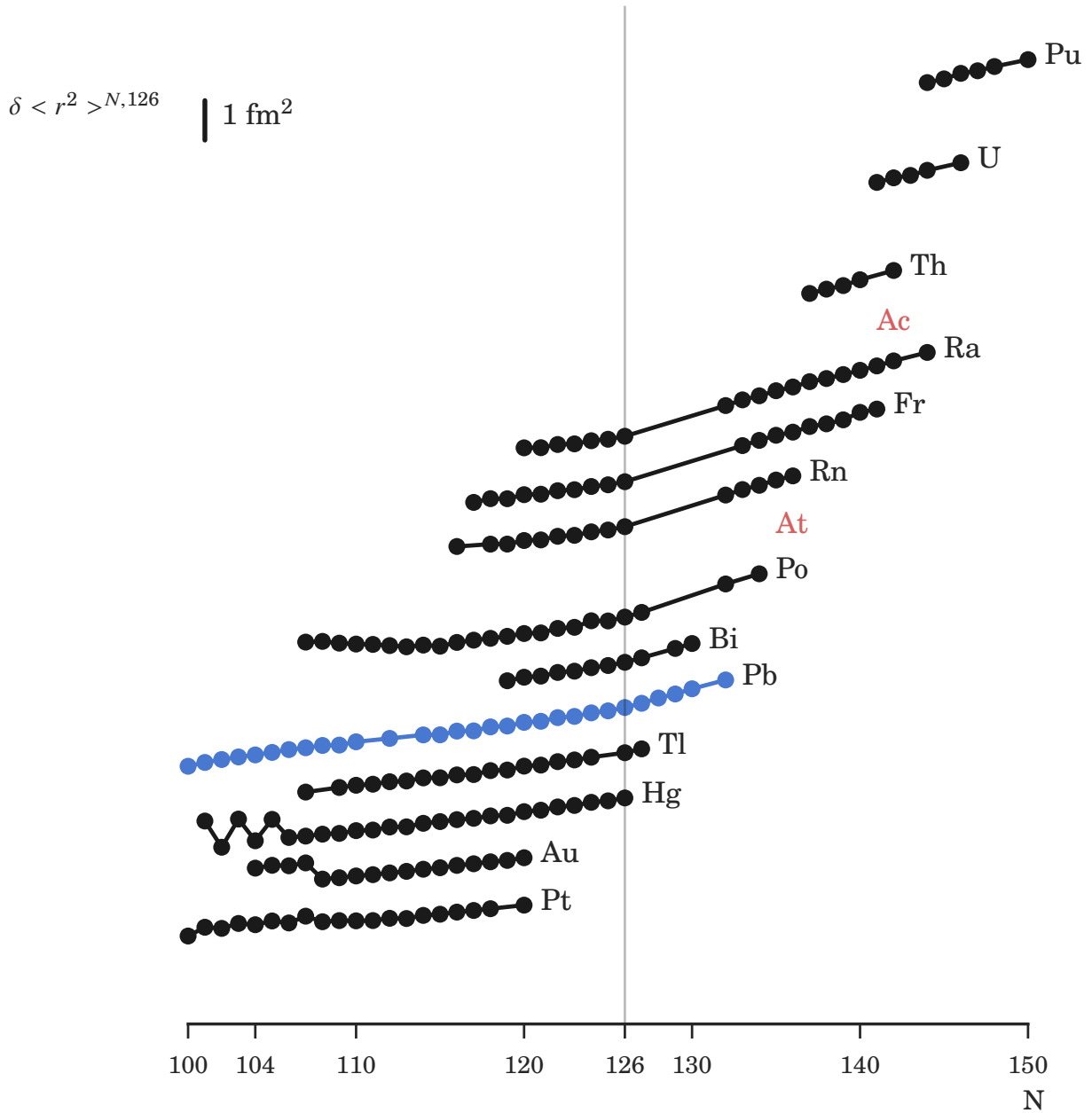


Figure 1.1: Change of the mean squared charge radii $\delta \langle r^2 \rangle$ around the lead isotope chain. In this mass region $\delta \langle r^2 \rangle$ is directly proportional to the IS. The datasets are separated by 1 fm^2 . The data were extracted from [11].

2 | Isotope production and detection

ISAC-I and ISAC-II Facility

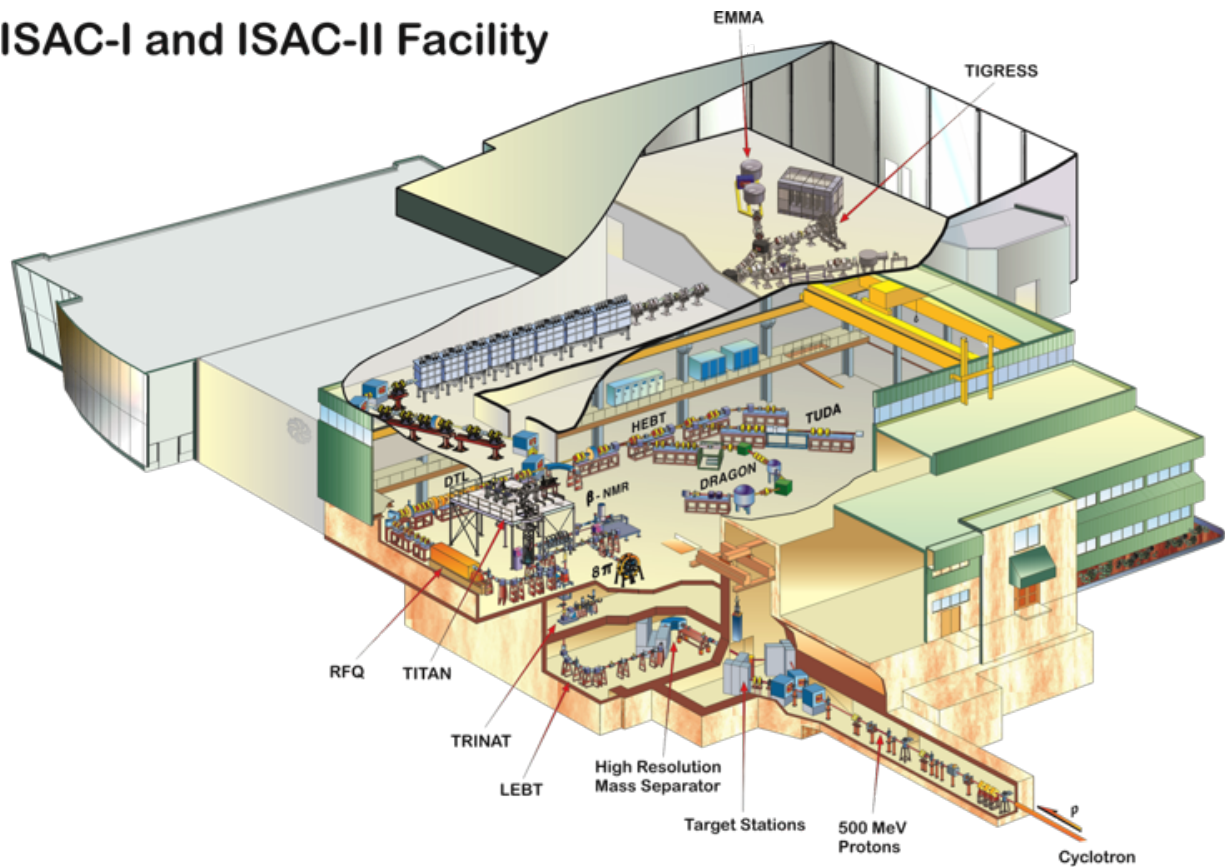


Figure 2.1: Schematic of the Isotope Separator and ACcelerator (ISAC) radioactive ion beam facility at TRIUMF [14].

RIBs at TRIUMF are produced at the ISOL facility ISAC shown in Figure 2.1. The primary beam is an up to $100\ \mu\text{A}$ proton beam provided by TRIUMF's 480 MeV H^- -cyclotron. This proton beam can be delivered to one of two ISAC target stations, named EAST and WEST after their relative locations in the target hall. The target container itself is a 20 cm long and 19 mm diameter tantalum

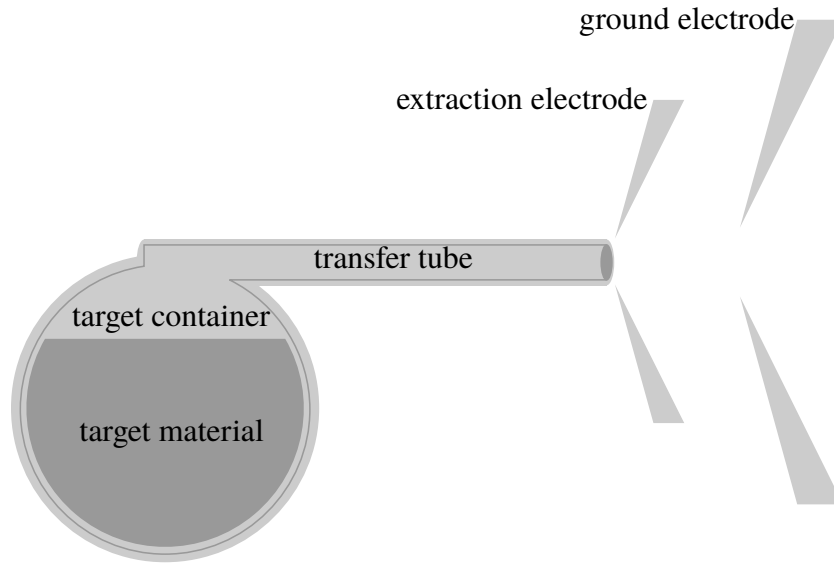


Figure 2.2: Schematic drawing of the surface ion source target module with two-stage ion beam extraction.

tube, containing a stack of D-shaped foils of the target material. A schematic drawing of the ISAC target can be found in Figure 2.2.

Target elements used at TRIUMF so far are silicon, titanium, nickel, zirconium, niobium, tantalum, thorium and uranium. The elements investigated in this work were extracted from either ThO or UC_x targets. Rare isotopes are produced by different nuclear reactions such as fission, fragmentation and spallation induced from the accelerated proton beam. Actinium and astatine are mainly spallation products. On the other hand, the production mechanism for silver is generally fragmentation. The ion beam intensity I_{ion} extracted from the target can be described by the following expression:

$$I_{\text{ion}} = \Phi \sigma N_{\text{target}} \epsilon, \quad (2.1)$$

where Φ is the proton beam intensity, σ is the reaction cross-section, N_{target} is the number of target atoms per surface area, and ϵ is the total efficiency. The latter can be described by the product of a series of partial efficiencies

$$\epsilon = \epsilon_{\text{release}} \epsilon_{\text{ion}} \epsilon_{\text{trans}}, \quad (2.2)$$

namely the release $\epsilon_{\text{release}}$, ionization ϵ_{ion} and transportation ϵ_{trans} efficiencies. In some cases, further terms need to be added such as cooling and bunching, charge-state breeding, and acceleration efficiencies [4]. Since for the experiments discussed in this work no post acceleration is necessary, only the efficiencies listed in eq. (2.2) will be further discussed.

2.1 Isotope release from the target

For ISOL the reaction products are stopped within the target material. The radioactive isotopes need to diffuse out of the target material and then effuse within the target container via a 3 mm transfer tube towards to the ionization region in order to be ionized and extracted from the target. This is realized by heating the target container to 2000 °C and above, where the temperature limit is given by the vapor pressure of the target material and the target container. A large part of the heating contribution comes from the power deposition from the proton beam which reaches up to 10 kW. In addition to this, the target container can be resistively heated with up to 600 A to compensate for lower proton beam currents [15]. Only a fraction of the isotopes produced in the target will actually make it to the experiment, as indicated in eq. (2.1). The first factor in eq. (2.2) is the release efficiency $\epsilon_{\text{release}}$ which describes the probability of a produced isotope reaching the ionizer before it decays. The release efficiency can be calculated by:

$$\epsilon_{\text{release}} = e^{-\frac{\tau_{\text{release}} \ln(2)}{t_{1/2}}}, \quad (2.3)$$

where $t_{1/2}$ is the half-life for each isotope and τ_{release} is the release time as defined by the mean time taken from the moment of production to the moment of extraction from the ion source. It is composed of

$$\tau_{\text{release}} = \tau_{\text{diffusion}} + \left(\tau_{\text{flight}} + \tau_{\text{adsorption}} \right) n, \quad (2.4)$$

where $\tau_{\text{diffusion}}$ is the diffusion time of the atom from the production origin to the surface, strongly depending on the target material density and chemistry [16]. The effusion process is defined by

the flight time τ_{flight} of the atoms between the collisions with the target and ion source walls. Since the flight time is typically $\tau_{\text{flight}} \leq 10^{-4}$ s, this process is negligible. Therefore the adsorption time $\tau_{\text{adsorption}}$ of the atom on the target and ion source wall, and $\tau_{\text{diffusion}}$, have the main impact on the release time. For a standard ISAC-target a total number of a few hundred thousand wall collisions n is expected before the atom is extracted from the ion source [17]. Release times for many elements are on the order of 1 s to 50 s [16], but in some cases can also require hours, days or in the case of refractory metals can even be infinite. Since transport times from the ion source to the experiment can be neglected, the release time is the main reason why the half-life of the isotopes produced with the ISOL method is limited. So far, the isotope with the shortest half-life delivered to an experiment at ISAC was ^{52}Ca with $t_{1/2} = 4.6$ ms [18].

Actinium, however, is an element with a long release time and is one of the elements to be investigated herein. During this work actinium was scheduled at TRIUMF three times for spectroscopy: once on a ThO- and twice on UC_x-targets. Whereas from a ThO-target no actinium could be extracted, it had been shown in previous experiments that from a UC_x-target weeks after the protons were turned off, the long-lived actinium atoms were still being released [19]. In order to estimate the release time from a UC_x-target¹, the yield for several isotopes was measured and divided by the theoretical in-source production rates [20] which results in the overall efficiency as shown in eq. (2.1). Now eq. (2.3) can be multiplied with the constant transport and ionization efficiencies and fitted to the data as shown in Figure 2.3. As a result, a release time of $\tau_{\text{release}} = 19(5)$ h and a constant efficiency of $\epsilon_0 = 0.5(1)$ % was extracted. Since the release time depends on many different factors, such as the density and composition of the target material and the target temperature, it is different for every target and can only be taken as an order of magnitude estimate. Considering a surface ionization efficiency of $\epsilon_{\text{ion}} = 0.85$ % as shown in Figure 2.4, the transportation efficiency can be determined to $\epsilon_{\text{trans}} = 58$ %. As for this particular target module, the extraction electrode got in contact with the target heat shield, the extraction electrode was shorted, which results in

¹ITW-TM1-UC#15-LP-SIS (ISAC-target station west - target module 1 - uranium carbide number 15 - low power - surface ion source).

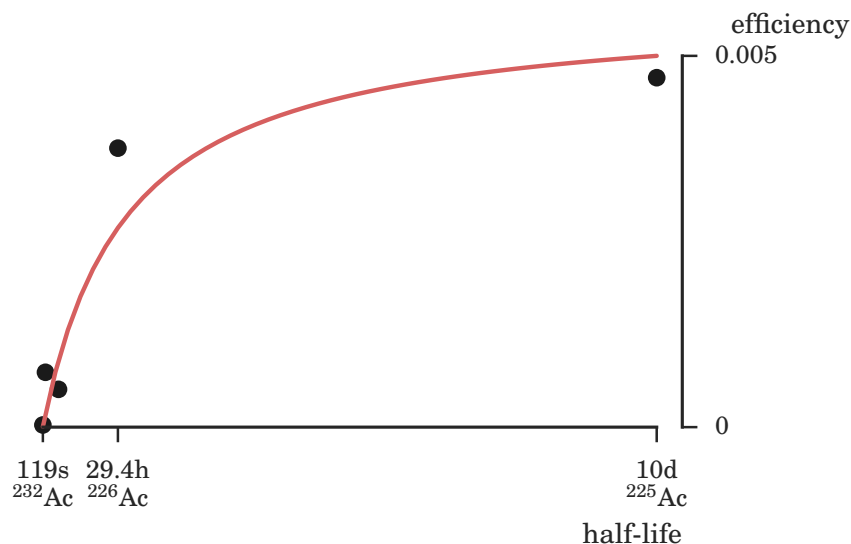


Figure 2.3: Actinium release from a UC_x target.

a reduced extraction efficiency. Therefore the result agrees very well with the expected transport efficiencies of 50 % to 70 % as discussed in section 2.3.

2.2 Ion sources

Once the isotopes are produced and have migrated from the target container to the transfer tube, the isotope of interest needs to be ionized for further mass separation, acceleration and transport. In contrast to the cyclotrons at TRIUMF which accelerate negative hydrogen ions, at the ISAC facility positive ions are extracted. Since no ion source in existence ionizes all elements with efficiencies $> 1\%$, at ISAC three different ion sources

- surface ion source (SIS),
- TRIUMF's resonant ionization laser ion source (TRILIS),
- forced electron beam induced arc discharge ion source (FEBIAD)

are available, each specialized for a specific group of elements. The elements thus far produced and ionized at ISAC are shown in Figure 2.5.

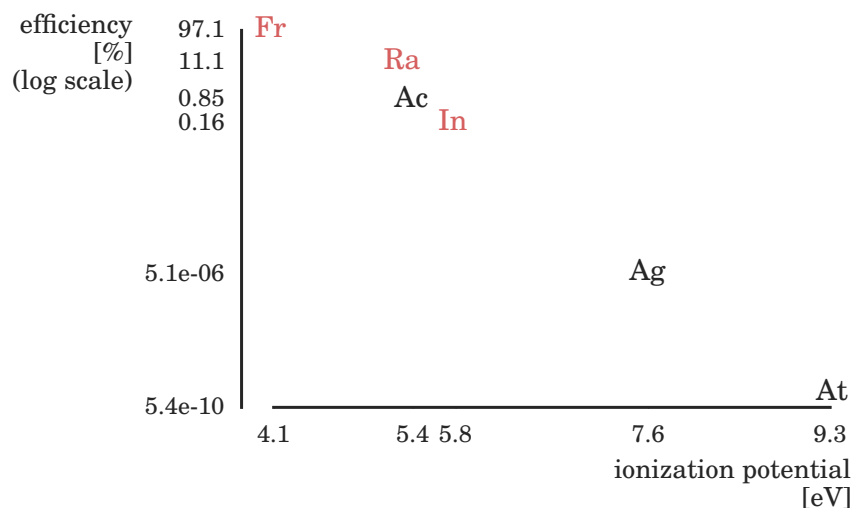


Figure 2.4: Theoretical surface ionization efficiency on a Re foil at 2000 °C for the elements of interest (labeled black) based on eq. (2.6). The elements that could cause isobaric contamination are labeled red. Francium and radium are part of the isobaric background for actinium and astatine, while indium can be found in a RIB of silver.

1 H 13.8																	2 He 24.6
3 Li 5.4	4 Be 9.3											5 B 8.3	6 C 11.3	7 N 14.5	8 O 13.6	9 F 17.4	10 Ne 21.6
11 Na 5.1	12 Mg 7.6											13 Al 6.0	14 Si 8.2	15 P 10.5	16 S 10.4	17 Cl 13.0	18 Ar 15.8
19 K 4.3	20 Ca 6.1	21 Sc 6.6	22 Ti 6.8	23 V 6.8	24 Cr 6.8	25 Mn 7.4	26 Fe 7.9	27 Co 7.9	28 Ni 7.6	29 Cu 7.7	30 Zn 9.4	31 Ga 6.0	32 Ge 7.9	33 As 9.8	34 Se 9.8	35 Br 11.8	36 Kr 14.0
37 Rb 4.2	38 Sr 5.7	39 Y 6.2	40 Zr 6.6	41 Nb 6.8	42 Mo 7.1	43 Tc 7.3	44 Ru 7.4	45 Rh 7.5	46 Pd 8.3	47 Ag 7.6	48 Cd 9.0	49 In 5.8	50 Sn 7.3	51 Sb 8.6	52 Te 9.0	53 I 10.5	54 Xe 12.1
55 Cs 3.9	56 Ba 5.2	*	72 Hf 6.8	73 Ta 7.5	74 W 7.9	75 Re 7.8	76 Os 8.4	77 Ir 9.0	78 Pt 9.0	79 Au 9.2	80 Hg 10.4	81 Tl 6.1	82 Pb 7.4	83 Bi 7.3	84 Po 8.4	85 At 9.3	86 Rn 10.7
87 Fr 4.1	88 Ra 5.3	**	104 Rf 6.0	105 Db 6.8	106 Sg 7.8	107 Bh 7.7	108 Hs ?	109 Mt ?	110 Ds ?	111 Rg ?	112 Cn ?	113 Nh ?	114 Fl ?	115 Mc ?	116 Lv ?	117 Ts ?	118 Og ?
*Lanthanides			57 La 5.6	58 Ce 5.5	59 Pr 5.5	60 Nd 5.5	61 Pm 5.6	62 Sm 5.6	63 Eu 5.7	64 Gd 6.1	65 Tb 5.9	66 Dy 5.9	67 Ho 6.0	68 Er 6.1	69 Tm 6.2	70 Yb 6.3	71 Lu 5.4
**Actinides			89 Ac 5.4	90 Th 6.3	91 Pa 5.9	92 U 6.2	93 Np 6.3	94 Pu 6.0	95 Am 6.0	96 Cm 6.0	97 Bk 6.2	98 Cf 6.3	99 Es 6.4	100 Fm 6.5	101 Md 6.6	102 No 6.7	103 Lr 4.9

Figure 2.5: Elements produced, ionized and extracted at ISAC. Selected is the most efficient ionization method for this element at the time of submission of this work. For many further elements, laser ionization would enhance the ionization efficiency, but ionization schemes have not yet been tested on-line at ISAC.

With the exception of TRILIS, the ion sources are directly attached to the target container. The target ion source combination is in operation for approximately four weeks and provides RIB for up to five different experiments in sequence. Since most experiments require the maximum ion beam intensity I_{ion} available, the target ion source combinations need to be wisely chosen for the scheduled experiments.

2.2.1 Surface ion source

Particles that get in contact with a hot surface have a probability that they desorb from the surface in a certain charge state. This effect can be used for positive as well as negative ion production and is known as surface ionization. Since ISAC operates usually with positive ions, only the first case will be considered further. At thermal equilibrium, the degree of ionization α is defined as the ratio of the number of ions N_+ to the number of neutral atoms N_n

$$\alpha = \frac{N_+}{N_n} = \frac{g_+}{g_n} e^{\frac{W-\text{IP}}{k_B T}}. \quad (2.5)$$

Equation (2.5) is called the Saha-Langmuir equation [21], where $g = 2J + 1$ is the statistical weight of the atom or ion, J is the total angular momentum of the state, W is the work function of the hot cavity material, IP is the ionization potential of the first ion, k_B is the Boltzmann constant, and T is the absolute temperature of the cavity. The ionization efficiency is given by [22]:

$$\epsilon_{\text{ion}} = \frac{\alpha}{1 + \alpha}. \quad (2.6)$$

Surface ionization is the most common ionization method for elements with low ionization potential such as the alkali metals, heavy alkali earth metals, lanthanides, and actinides, due to its simplicity and high efficiencies. For the alkali metals, ionization efficiencies up to 100 % can be achieved. The surface ion source is the simplest design as it consists only of the heated tantalum transfer tube. In order to increase efficiency, the source should be operated at high temperatures using

materials having high work functions. For this reason, a thin rhenium foil with a work function of $W = 4.8$ eV is inserted into the transfer tube. The calculated ionization efficiencies at $T = 2000$ °C for the elements of relevance to this work are shown in Figure 2.4.

2.2.2 TRILIS

Laser ion sources are based on the stepwise excitation of a valence electron into the continuum. In general, one to two resonant transitions into intermediate excited states are followed by the final ionizing step, which can be realized either nonresonantly by a high power laser or resonantly into a Rydberg or autoionizing state (AI); more details can be found in section 3.2. Since the excitation schemes vary between different chemical elements, these ion sources ionize element-selectively. They are most suitable for elements with ionization potentials between 5 eV to 9.5 eV. With resonant laser ionization, efficiencies in excess of 20 % have been accomplished [23].

Laser beam transport

In order to match the laser frequency to the transition frequency of the atom, a set of tunable lasers is required. The TRILIS on-line laser laboratory is equipped with four birefringent filter (BRF)-tuned titanium-doped sapphire (Ti:Sa) lasers, one ionization laser for nonresonant ionization, and two frequency-tripling/quadrupling units. The Ti:Sa lasers are pumped with one frequency-doubled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser. All of these components are discussed in detail in chapter 4.

The laboratory is located on the floor of the low energy experimental hall in ISAC and therefore two floors above the target stations. This has the advantage that all operational parts of this ion source are away from any radiation. In order to get the laser light to the ionization area which is the transfer tube at the target container, all lasers with similar wavelengths are combined on the laser table and transported via dielectric mirrors down to the mass separator area. Due to the

natural divergence of a laser beam and the long transport path from the on-line laser laboratory to the ionization region of about 20 m, the lasers have to be focused in order to match the laser diameter with the transfer tube diameter of 3 mm. Depending on which target station is in use, the laser beams are transported to one of two laser tables where the focusing lenses are located and the beams are overlapped using dichroic mirrors. These laser tables are installed approximately 9 m away from the ionization region. In order to obtain the beam diameter in the transfer tube of < 3 mm despite the long focal length, the laser beams are expanded by a factor 3 to 5 with Galilean type telescopes in the laser laboratory. The laser light accesses the ionization region via a fused silica vacuum window located at the pre-separator vacuum vessel. A reflection off this window acts as a reference indicating whether all lasers are focused and overlapped in the source. The reference spot is monitored via a camera on a ceramic disc². This disc is placed at the same distance to the window as it is to the transfer tube, about 4 m to 6 m depending on the setup [24]. A schematic drawing on the laser beam transport is shown in Figure 2.6.

Ion guide laser ion source

During normal operation, TRILIS is used in combination with the surface ion source target module. As a result, even though TRILIS ionizes elements selectively, other elements with low ionization potential are still contained in the RIB. For many experiments this results in an unacceptably high background for the desired measurements, even after mass separation in the ISAC mass separator. For that reason a target ion source has been designed to suppress those unwanted surface ions.

For the purpose of creating isobar free RIBs, the transfer tube is shortened and replaced by a cold radio frequency quadrupole (RFQ). Since the target and hence also the transfer tube are still heated to approximately 2000 °C to ensure fast release from the target, a significant number of surface ions are created in the transfer tube region. To prevent those ions from leaving the target and entering the ionization region, an additional repeller electrode is placed between the transfer tube and the

²Kentek ViewIt-IR.

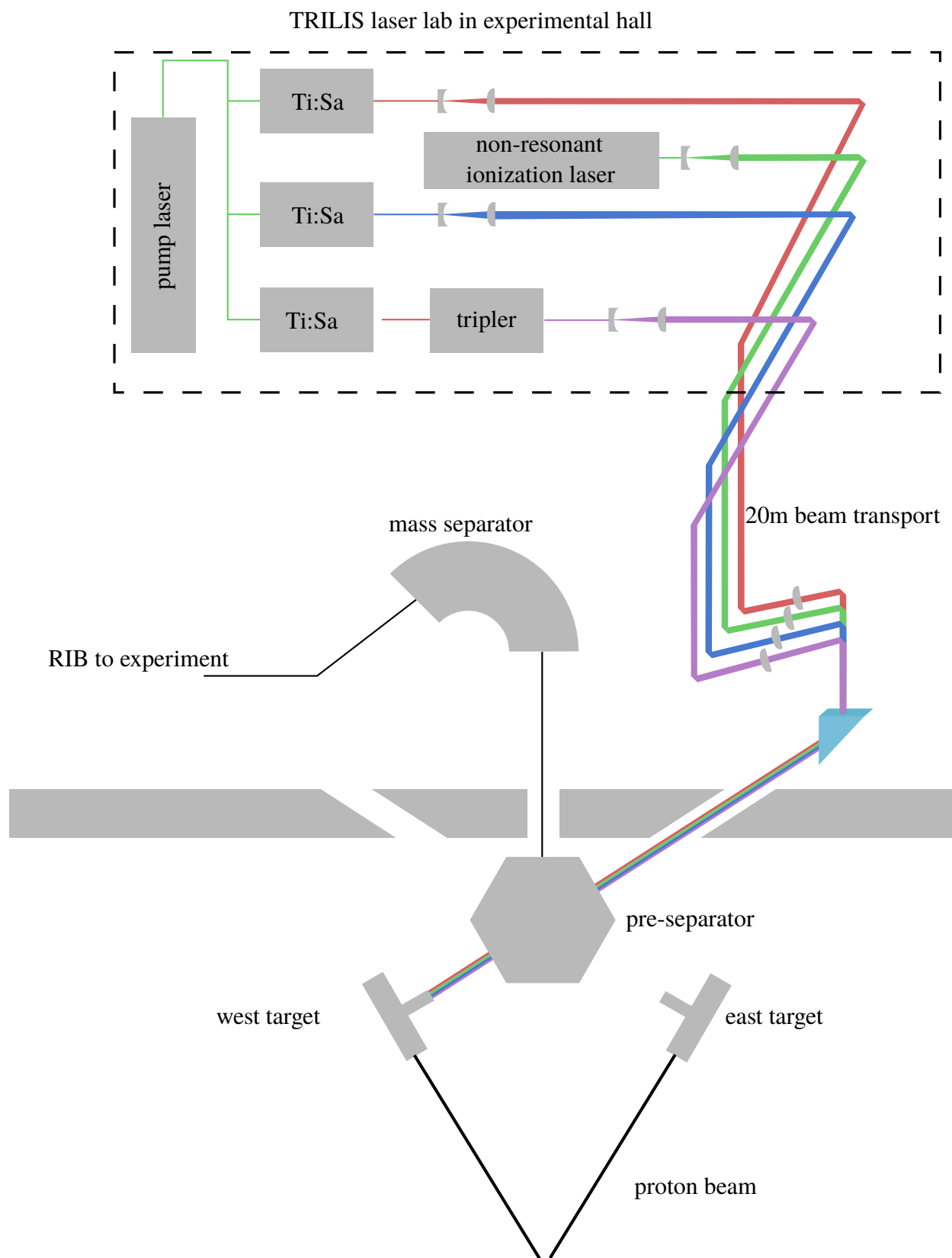


Figure 2.6: Laser beam transport to the west target station.

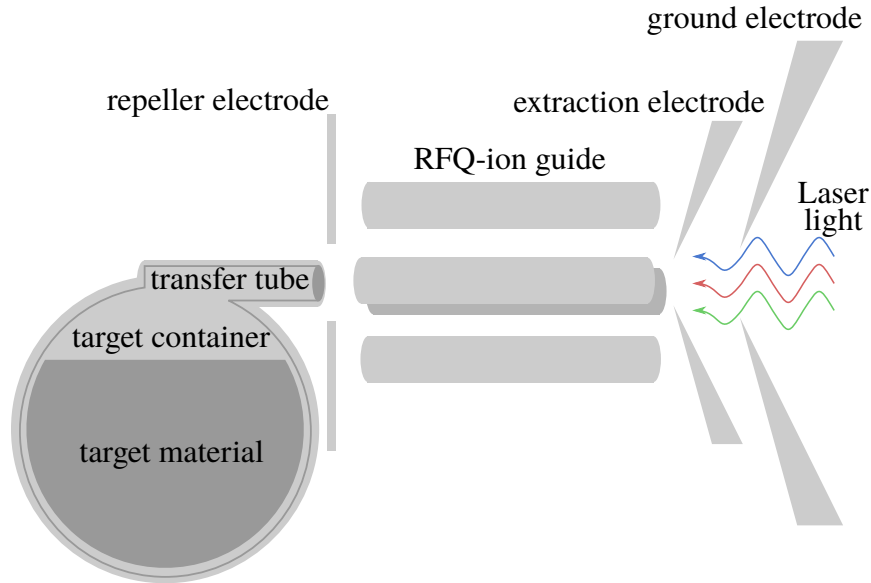


Figure 2.7: Schematic drawing of an IG-LIS target module with two stage extraction.

RFQ. By applying a voltage to the repeller electrode that is higher than the source bias, only neutral atoms are able to enter the ionization region where they can be ionized by the resonant laser light. However, by applying a repeller voltage smaller than the source voltage to the electrode, it is still possible to extract those surface ions. This is especially of interest for beam tuning purposes, or if experiments are scheduled that require surface-ionized RIBs. Once ionized, the RFQ prevents the ions from escaping the ionization region, and guides them towards the extraction electrode. A schematic drawing of this setup is shown in Figure 2.7.

With this setup, the ion guide laser ion source (IG-LIS) reduces surface-ionized isobaric background by up to six orders of magnitude. Compared to the laser ionized yield with a surface ion source module, the IG-LIS yields drop approximately by a factor of 50 [25].

2.2.3 FEBIAD

The FEBIAD is the only ion source able to ionize all elements. Common ionization efficiencies for this source are around 10 % [15], which is lower than the efficiencies that can be reached with the ion sources described above. Therefore, this source is only used for elements that cannot be

ionized by surface or laser ionization, namely, halogens, noble gases, or certain molecules such as CO. Furthermore, since all elements are ionized simultaneously, a high isobaric background is expected which in most cases is not desired by the experimenters.

2.3 Ion beam transport

Following the ionization process, the ions need to be transported to the experiment. Therefore the transfer tube is set between 12 kV to 60 kV, the acceleration voltage of the ion. The RIB is extracted by an extraction electrode set to approximately 10 % of the target bias. A ground electrode realizes the final acceleration as it introduces the largest potential step to that of the beamline at 0 V. The beam is transported by a set of steerers, an einzel lens and matching quadrupoles to a pre-separator which accomplishes multiple purposes, serving as a:

- mass separator with an achieved mass resolution $m/\Delta m > 200$ [26],
- switchyard to select the RIB from either target station, and,
- entrance for the laser light to be sent into the target.

Next, the RIB is sent to a mass separator with a measured resolving power of 2000 [27] which selects the final mass of interest. Finally the selected isotope can be sent to an ion detector, the Yield Station, or the requesting experiment. The transport efficiency ϵ_{trans} describes the losses related to the beam transport from the ion source to the experiment. This includes extraction efficiencies and transmission through the pre- and mass separator. While the transmission through the mass separator is on the order of 90 % [26], the overall transmission efficiency cannot be directly measured: in front of the pre-separator all ions extracted from the ion source are included, whereas behind the mass separator the RIB only contains the requested mass. The overall transmission efficiency at ISAC is estimated between approximately 50 % and 70 % [28].

2.4 RIB detectors

Depending on the intensity of the RIB, different detectors can be used to measure the ion current.

2.4.1 Faraday cup

The simplest detector is a Faraday cup (FC) which, in principle is a metal cup directly connected to an ampere meter. Charged particles that impinge on the metal are absorbed and the resulting current can be measured. By preventing that secondary electrons which are created at the metal surface escape the FC, the measured current directly corresponds to the beam intensity. The detection limit for FCs at ISAC is at approximately 1 pA, which corresponds to 6.3×10^9 counts per second (cps). Since FCs accept high ion currents these detectors are often used as beam dumps along the low energy beamlines at ISAC.

2.4.2 Channeltron electron multiplier

For low intensity RIBs, channeltron electron multipliers (CEMs) are used for beam diagnostics. In this detector, the impinging particle creates secondary electrons triggering an avalanche of further secondary electrons thereby creating measurable currents. CEMs have a maximum detection limits of 10^6 cps. In order to protect the CEMs from high count rates, it is possible to place attenuators in the ion beam path. The attenuators consist of perforated metal sheets where the hole density corresponds to the attenuation factor. Currently two attenuators with factors of 10 (real attenuation 7) and 100 (real attenuation 60) are available. The CEM used in this work is CEM 20, which is located approximately 5 m downstream of the high resolution mass separator.

2.4.3 TRIUMF Yield Station

Many experiments based at the ISAC facility require a specific RIB purity and intensity in order to be conducted. Before an RIB can be delivered to an experiment, the content of the beam has to be characterized for these needs. This is realized at the ISAC Yield Station [29], a nuclear detection chamber located in the ISAC experimental hall at the end of the vertical beamline section. The Yield Station contains an aluminized Mylar[®] tape³ where the radioactive isotopes are implanted for a specified collection time. After implantation, a fast kicker steers the RIB to a FC (acting as a beam dump) located in the mass separator room directly behind the mass separator. The implanted isotopes are allowed to decay and the resulting α -, β -, and γ -emission can be simultaneously identified by three different types of detectors, each specialized for one type of radiation. A schematic drawing of the Yield Station interior, and a photo of the vacuum chamber with the high-purity germanium detector (HPGe) is shown in Figures 2.8 and 2.9.

- Four plastic scintillators⁴ convert the kinetic energy of a β -particle into light which can be detected by a photomultiplier. Depending on the size and location of the scintillator the detection efficiency varies between 0.5 % to 48 %. The power supply of each photomultiplier can be independently controlled to regulate the overall efficiency, in addition to varying the collection time and protecting the photomultiplier from over-current damage.
- Two windowless PIN diodes⁵ are placed at a 45° angle underneath the implantation spot on the tape to detect α -particles. The maximum detection efficiency of each diode is about 0.5 %. An optional third diode can be moved directly into the RIB which increases the efficiency to close to 50 %. Since all nuclei must have decayed before the new implantation cycle starts, this option is only suitable for RIB with short-lived isotopes.

³A polyester foil made of polyethylene terephthalate with high tensile strength, chemical and dimensional stability and electrical insulation.

⁴Saint-Gobain BC-408.

⁵Hamamatsu S3590-9, surface area 100 mm².

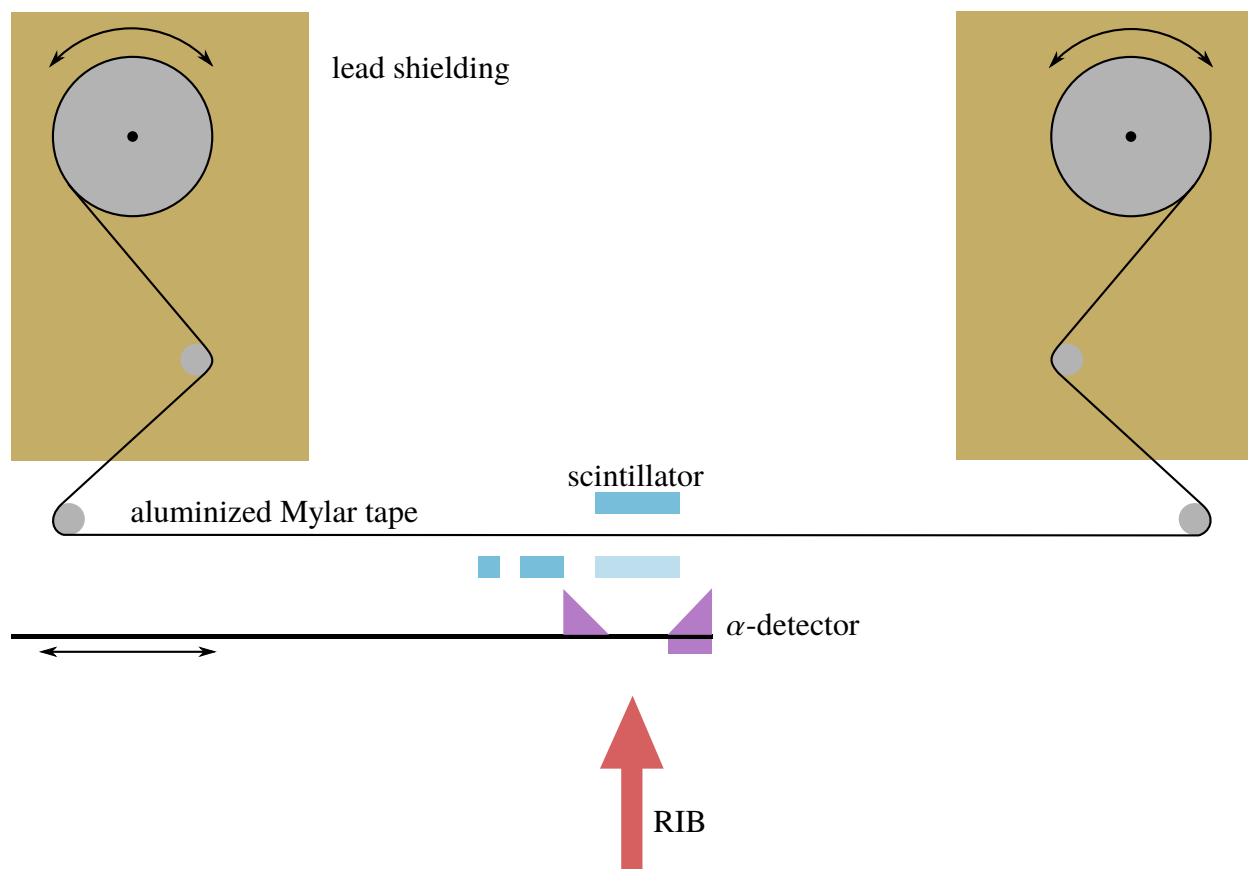


Figure 2.8: Schematic drawing of the detector system inside the ISAC Yield Station vacuum chamber. The radiation of the implanted RIB can be detected by three different sets of detectors. A set of scintillators (blue) is able to detect the emitted β -radiation. The scintillator labeled light blue is spatially displaced in order to allow the RIB to reach the tape. The emitted α -particles are detected by windowless PIN diodes (indicated in purple) mounted on a movable rail. In normal operation the two diodes mounted at a 45° angle to the rail are used. For low-intensity RIBs with short $t_{1/2}$ it is possible to implant the RIB directly into a PIN photo diode mounted on the bottom of the rail. The germanium detector for measuring γ -radiation is located outside the detection chamber and shown in Figure 2.9.

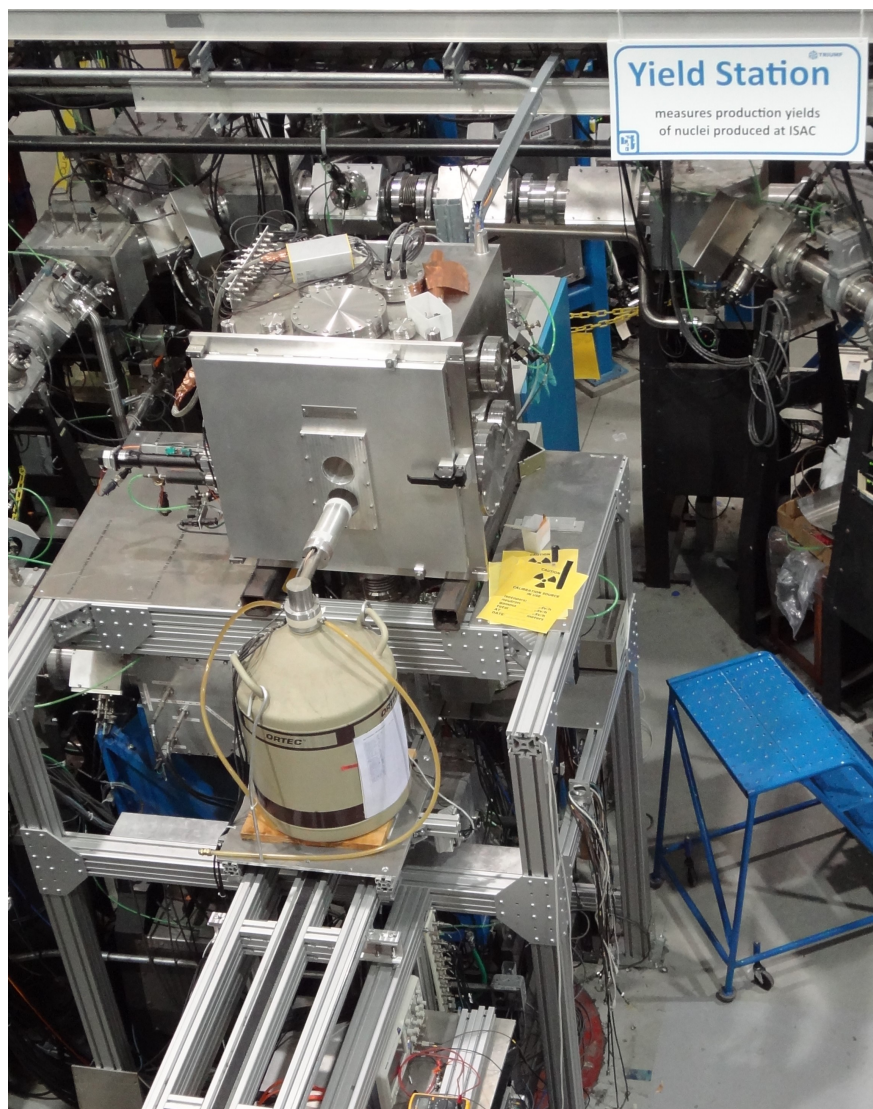


Figure 2.9: Yield Station detection chamber with the rail mounted HPGe detector and its liquid nitrogen dewar.

- A HPGe⁶ is placed on a rail outside the vacuum chamber for γ -radiation detection. The efficiency of a germanium detector is photon energy-dependent and has its maximum at approximately 150 keV [30]. By changing the distance between the vacuum chamber and the detector, the solid angle detection efficiency can be varied by two orders of magnitude.

At the end of a measurement cycle, the irradiated part of the tape is moved behind lead shielding and the process can be repeated on a fresh piece of tape. For the tape to move and to recover its tension, a time period of 4 s is set before each new implantation cycle. The number of repetition cycles, and collection and decay time can be chosen depending on the half-life of the isotope of interest, the expected intensity of the beam, and possible contamination.

The yields for α - and γ -emitting isotopes are determined by counting the events with corresponding photon energies. For the final determination, the branching ratio of the decay and the detector efficiency have to be incorporated. For β -emitters, only the number of events in sequential time intervals (dwell time) is recorded. In this case, a set of decay equations is fitted to the experimental decay curve. Subsequently the yield is determined from the number of events in the first time bin.

In addition to determining yields in preparation for upcoming experiments and characterizing the target and ion source performance, the Yield Station can also be used as a diagnostic tool for resonance ionization laser spectroscopy; which is the setup mainly used in this work. For this purpose, the etalon of the laser to be scanned is equipped with a piezo motor that can be remotely controlled by the Yield Station software. With each new implantation cycle, the piezo motor moves a specified number of steps which changes the wavelength of the laser light by tilting the etalon. Especially for short-lived isotopes, the number of decay events per wavelength step are therefore limited with the common counting setup. However, since for this kind of measurement the actual yield is not of interest, the counting setup is changed such that it also counts events in the collection time (as opposed to only during the decay time as is done for yield measurements). Thus the counting statistics can be increased by increasing the collecting time. The advantage of using the

⁶Ortec GMX40, 40 % rel. efficiency.

Yield Station compared to an ordinary ion counting detector such as a channeltron or Faraday cup is that laser frequency dependence can be studied for each component of the RIB independently. This is especially of interest for RIB with high isobaric background, separation of ground and isomeric states of an isotope, or assignment of γ - or α -energies to a certain isotope.

3 | Resonant laser ionization

3.1 Excitation scheme selection

Each chemical element has an individual atomic energy level structure which arises from the quantum mechanical properties and Coulomb interaction between the electrons and the atomic nucleus. Spectroscopic characteristics allow for element selectivity in resonant ionization laser ion sources. The efficiency of a laser ion source strongly depends on the chosen excitation scheme. Electrons in an atom can be excited into a higher energy state if the photon energy $h\nu_{ik}$, where h is the Planck's constant and ν is the photon frequency, matches the energy difference ΔE between the lower state $|k\rangle$ and an excited state $|i\rangle$

$$h\nu_{ik} = |E_i - E_k| = \Delta E. \quad (3.1)$$

Based on a single-electron atom, every electronic state can be fully described by a set of quantum number $nlm_l m_s$. The principal quantum number n is a positive integer, and the orbital angular momentum $l = 0, 1, 2, \dots, n - 1$ where in this case every number is associated with a letter ($0 = s$, $1 = p$, $2 = d$, $3 = f$, $4 = g, \dots$). The magnetic quantum numbers m_l and m_s are the projection of the orbital angular momentum l and the intrinsic spin s of the electron on an arbitrarily chosen z -axis of the atom. Therefore the magnetic quantum number for the angular momentum can have numbers between $m_l = -l, -(l - 1), \dots, (l - 1), l$ and the magnetic spin number is $m_s = \pm 1/2$.

3.1.1 Selection rules

Even if the condition in eq. (3.1) is fulfilled, not all transitions between existing energy levels are allowed since in addition to energy, angular momentum also has to be conserved. Since photons carry an angular momentum of $\pm\hbar$, electric dipole transitions are allowed only where

$$\Delta l = \pm 1 \quad (3.2)$$

and spin remains unaffected

$$\Delta s = 0. \quad (3.3)$$

These selection rules apply strictly only in light elements where the spin-orbit coupling is weak. For heavier elements, the electron spin and orbital angular momentum couple to a total angular momentum $\vec{j} = \vec{l} + \vec{s}$. For these atoms, the most interesting quantum number is the total angular momentum J . Depending on the mass of the nucleus, J is treated differently; the two extremes are spin-orbit coupling and $j - j$ coupling.

The spin-orbit coupling is valid for light elements; here, $\vec{J} = \vec{L} + \vec{S}$ where $\vec{L} = \sum_i \vec{l}_i$ and $\vec{S} = \sum_i \vec{s}_i$. For these elements, eqs. (3.2) and (3.3) are also valid for the total spin S and total orbital momentum L . From these rules it can be seen that the selection rule for the total angular momentum J is

$$\Delta J = 0, \pm 1 \quad \text{but} \quad J = 0 \rightarrow J = 0, \quad (3.4)$$

since only the magnitude of S in eq. (3.3) is important, not the direction.

In order to more quickly see whether a transition is allowed, the electron configuration of an energy level is typically given in the so-called spectroscopic notation

$$^{2S+1}L_J, \quad (3.5)$$

where the term $2S + 1$ describes the multiplicity or the number of fine structure components of the level, and L is expressed in capital letters similar to those for the individual orbital angular momentum l as mentioned above.

With increasing proton number Z , the spin s and orbital momentum l of each electron couple first to a single total angular momentum j_i . The total angular momentum of the atom is then formed by $\vec{J} = \sum_i \vec{j}_i$. Even though the L-S coupling is no longer valid for these elements, the electron configuration is often still expressed as shown in eq. (3.5). It should be noted that the selection rules expressed in eqs. (3.2) and (3.3) cannot be applied anymore in this case. The only significant selection rule is then eq. (3.4).

Energy levels and allowed transitions including corresponding transition strengths for most elements are tabulated in databases [8, 9]. Especially for transitions from higher excited states, these databases are incomplete so that further spectroscopy is necessary in order to find the combination of the strongest transition for the most efficient ionization schemes.

3.1.2 Thermal population

The isotopes of interest are produced and ionized in a hot environment, typically at temperatures around $T \approx 2000$ K. At these temperatures it is possible that low lying excited states can be thermally populated and significantly decrease the population of the atomic ground state. For atoms in a thermal equilibrium, the ratio between the number of atoms in the ground state N_0 and the number of atoms N_i in an excited state E_i can be described by the Boltzmann distribution

$$\frac{N_i}{N_0} = \frac{g_i}{g_0} e^{-\frac{E_i - E_0}{k_B T}}, \quad (3.6)$$

where k_B is the Boltzmann constant and $g_i = 2J_i + 1$ is the level of degeneracy of the state E_i . For the elements covered in this work, the only element with an low-lying excited state of relevance is actinium. It has an excited state of $E_1 = 2231.43 \text{ cm}^{-1}$ with a total angular momentum $J_i = 5/2$

and the ground state has total angular momentum $J_0 = 3/2$. Consequently, at the temperature of operation, approximately 30 % of the atoms occupy the low-lying excited state.

3.2 Ionization process

After selecting the resonant steps to excite the electron into a higher energy level, the final ionizing step has to be chosen. This can be realized by three different methods:

- nonresonant ionization into the continuum,
- resonant ionization into an autoionizing state above the ionization potential, or
- electric-field or far-infrared photoionization after a resonant excitation into a high lying Rydberg state

as illustrated in Figure 3.1, which will be further discussed in detail in the following sections.

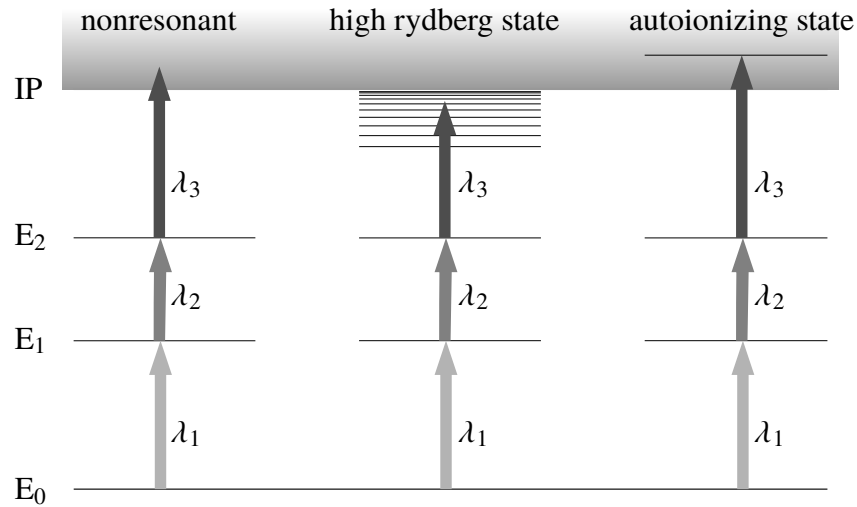


Figure 3.1: Ionization processes for laser resonant excitation schemes. The number of resonant excitation steps before ionization of an element varies. The ionization step is usually the rate-limiting process and therefore requires special attention. Moreover, for most elements the knowledge of the autoionizing state spectrum is limited despite the often observed large transition strengths.

3.2.1 Nonresonant ionization

Nonresonant ionization is the most straightforward photoionization method. The only condition that needs to be fulfilled is for the photon energy of the ionization laser $h\nu$ to be equal or greater than the energy difference between the excited state E_k and the ionization potential (IP)

$$h\nu \geq \text{IP} - E_k. \quad (3.7)$$

In many cases, the photon energy of one of the excitation steps can be used. Although the wavelength of the ionization laser is not as critical as that for resonant transitions, the ionization cross-section decreases rapidly with increasing photon energies. Therefore, the highest ionization cross sections can be reached when the photon energy exactly matches the energy gap in eq. (3.7). Typical ionization cross-sections for nonresonant ionization are rather low: $\sigma_I \sim 10^{-17} \text{ cm}^2$ to 10^{-19} cm^2 [31, 32]. To achieve maximal ionization, the excited electron needs to be moved to the continuum before it relaxes. This can be obtained for $P_I > A_{ki}$ where P_I is the ionization probability and A_{ki} is the Einstein coefficient for spontaneous emission. The ionization probability for nonresonant ionization is given by

$$P_I = \sigma_I n(\nu) \quad (3.8)$$

where $n(\nu)$ is the photon flux [31]. For an excitation scheme with two resonant transitions, the lifetime of the highest excited energy level is in most cases $\tau \approx 10^{-7} \text{ s}$. It follows that the best-case photon flux for maximum nonresonant ionization is $n(\nu) > A_{ki}/\sigma_I = 10^{24} \text{ cm}^{-2} \text{ s}^{-1}$. For the Ti:Sa laser system running at 800 nm with a pulse length of 50 ns and a repetition rate of 10 kHz focused down to a spot size of 3 mm, the laser would have to deliver an output power of $P_{\text{out}} > 8.7 \text{ W}$. Since the output power from the Ti:Sa lasers is around 2 W to 3 W, at TRILIS an additional ionization laser is available.

3.2.2 Rydberg states

Excitation energies in highly excited atoms can be generally described by the Rydberg-Ritz formula:

$$E = \frac{Ry}{(n - \delta)^2}, \quad (3.9)$$

where Ry is the element-specific Rydberg constant, n is the principle quantum number, and δ is the quantum defect. The quantum defect is a shielding constant and a measure of how much the core electrons shield the Rydberg electron from the interaction with the nucleus. As hydrogen only has one electron, the quantum defect for this element is defined as $\delta = 0$ [33]. The quantum defect δ strongly depends on the probability of the electron to be found at a distance r away from the nucleus, which increases with increasing orbital quantum number l and decreasing atomic number Z [34].

As the energy difference from the IP decreases for highly excited Rydberg states, low energies such as provided by thermal photons, collisions with other atoms, or an external electric field can be enough to ionize the atom. The closer the excited electron gets to the IP, the more likely it will be ionized. A typical Rydberg series is shown in Figure 3.2. It can be seen that the states of interest have principle quantum number $n = 10$ to 40. The ionization cross-sections for these levels are around $\sigma_I = 10^{-12} \text{ cm}^2$ [23]; hence, approximately five magnitudes higher than for nonresonant ionization. During the spectroscopy on calcium where this Rydberg series was measured [35], it was possible to saturate a Rydberg transition with about 500 mW of laser power. Nevertheless, excitation schemes that uses Rydberg states as the ionizing transition are the least often schemes applied at TRILIS.

3.2.3 Autoionizing states

Up to now it was assumed that the photons involved in the multi-step excitation are absorbed by only one valence electron while the remaining electrons stay in their ground state. However, for

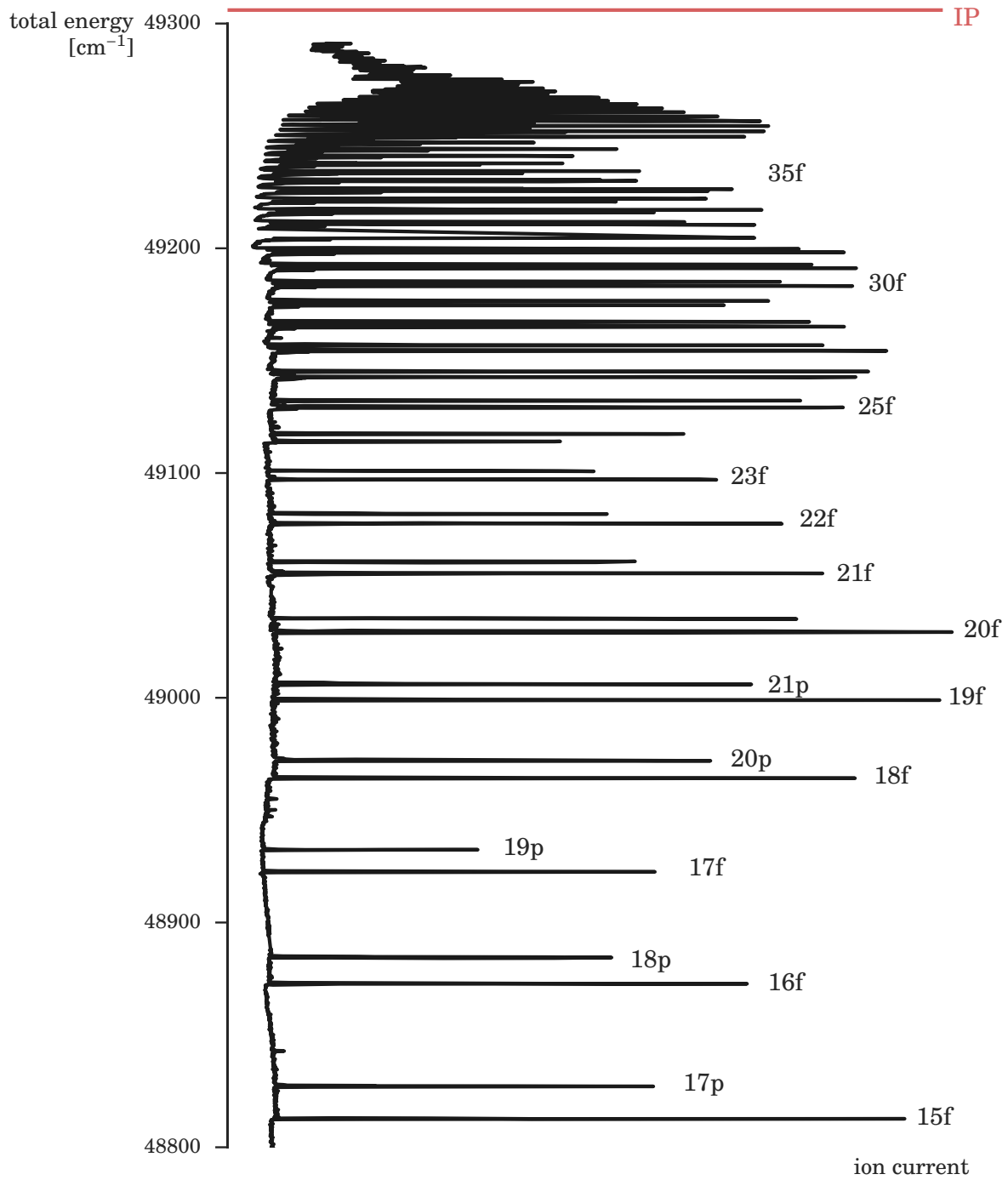


Figure 3.2: Rydberg series of calcium measured with the TRILIS laser system in the off-line development laboratory. Details of the excitation scheme applied can be found in [35].

atoms with multiple electrons it is also possible that two electrons can be excited simultaneously. Once the electrons are excited, both electrons can either decay by emitting two photons, or one of the electrons can transfer its excitation energy to the second excited electron which can lift it above the ionization potential. The latter process is called autoionization [34].

A characteristic of these resonances is an asymmetric lineshape, known as a Fano-Beutler profile, arising from interference between the nonresonant and multi-electron excitation ionizations [34, 36]. Autoionizing states typically have extremely short lifetimes on the order of $\tau = 10^{-13}$ s [37] leading to natural linewidths $\Delta\nu > 1600$ GHz. Typical ionization cross-sections for autoionizing states are $\sigma_I = 10^{-16} \text{ cm}^2$ to 10^{-15} cm^2 . For select resonances, cross-sections up to $\sigma_I = 10^{-12} \text{ cm}^2$ have been measured [38].

3.3 Broadening mechanisms

An electron exists in an excited state E_k for an average time τ , varying from a few ns for fast transitions up to several years for highly forbidden transitions. For that reason, due to the uncertainty principle

$$\Delta E \tau \geq \frac{h}{2\pi} \quad \text{with} \quad \Delta E = h\Delta\nu, \quad (3.10)$$

light absorbed or emitted from an atom will always have a minimum linewidth $\Delta\nu$, defined by the full width at half maximum (FWHM) $\Delta\nu \geq \frac{1}{2\pi\tau}$, called the natural linewidth. Since the lifetime of an excited state is the same for all atoms in an ensemble, the line broadening is homogeneous which leads to a Lorentzian line profile. Measuring an energy level with a precision near its natural limit requires some experimental effort [31, 34]. In general, additional broadening mechanisms are involved in the measurement. The broadening mechanisms of relevance for the here applied experimental setup will be described in the following sections.

3.3.1 Doppler broadening

The light absorbed by an atom moving relative to the light source with velocity $\vec{v}$ is shifted from its eigenfrequency ν_0 depending on the direction. For an atom moving in the z -direction with $\vec{v} = (0, 0, v_z)$, the light absorbed by the atom in the non-relativistic case is given by

$$\nu = \nu_0 \left(1 + \frac{v_z}{c} \right), \quad (3.11)$$

where c is the speed of light. This frequency shift is called the Doppler effect. Light absorbed by an atom moving towards the source is shifted to a shorter wavelengths, while an atom moving away from the source absorbs light towards a longer wavelengths.

The atoms to be ionized in the discussed experiments are in a low-pressure gas phase in which the atoms' motion can be described by a Maxwell-Boltzmann velocity distribution $n(v)$, given by:

$$n(v) = \sqrt{\frac{2}{\pi}} \left(\frac{m}{k_B T} \right)^{3/2} \cdot v^2 \cdot e^{-\frac{mv^2}{2k_B T}}, \quad (3.12)$$

where m is the mass of the atoms, k_B is the Boltzmann constant, and T is the absolute temperature of the ensemble. Since the intensity distribution $I(\nu)$ of the absorbed light is proportional to the velocity distribution $n(v)$ of the atoms, it can be seen in Figure 3.3 that this is larger for lighter atoms. By substituting eq. (3.11) in eq. (3.12), the intensity distribution can be expressed as

$$I(\nu) = I(\nu_0) e^{\left(\frac{\nu - \nu_0}{\nu_0} \frac{c}{v_p} \right)^2}, \quad (3.13)$$

where $v_p = \sqrt{2k_B T/m}$ is the velocity with the highest probability. The linewidth of this peak is

$$\Delta\nu_D = \frac{2\nu_0}{c} \sqrt{\frac{8k_B T \ln 2}{m}}. \quad (3.14)$$

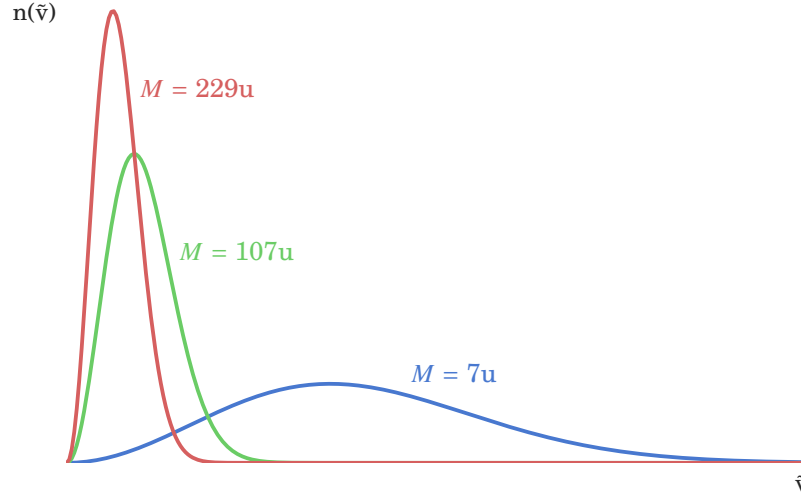


Figure 3.3: Maxwell-Boltzmann distribution for particles at constant temperature with atomic weights $M = 7 \text{ u}$, 107 u and 229 u . This distribution is responsible for the observed Doppler-broadened linewidth in in-source laser resonance ionization spectroscopy.

In order to simplify this expression, the Avogadro constant N_A can be introduced [31] allowing m to be replaced by the atomic weight $M = mN_A$, and k_B can be replaced by the gas constant $R = k_B N_A$. Equation (3.14) can then be written as:

$$\Delta\nu_D = \frac{\nu_0}{c} \sqrt{\frac{2RT \ln 2}{M}} = 7.16 \times 10^{-7} \nu_0 \sqrt{\frac{T}{M}} \left[\text{s}^{-1} \right]. \quad (3.15)$$

For the example of the first transition in silver, the Doppler broadening can be calculated. The temperature of the atomic vapor is typically 2000 K. Taking the first transition in silver shown in Figure 5.1, $\nu_0 = 913.55 \text{ THz}$. The most abundant stable isotope is ^{107}Ag . It follows that the Doppler width for this transition is $\Delta\nu_D = 2.8 \text{ GHz}$. The first excitation step in silver has a transition strength of $A_{ki} = 1.38 \times 10^8 \text{ s}^{-1}$ [9]; consequently, the natural linewidth of this transition is $\Delta\nu_n = A_{ki}/2\pi = 22 \text{ MHz}$. Hence the Doppler width in this case is about two orders of magnitude larger than the natural linewidth. For that reason the lasers applied at TRILIS have a linewidth of 1 GHz to 3 GHz in order to ionize the atoms in all velocity classes.

3.3.2 Saturation broadening

The saturation of an optical transition is essential for efficient laser ionization. If all transitions are saturated, the majority of atoms would be ionized with this ionization scheme, the ion current increase from applying more laser power would be negligible. In this work, the saturation power P_{sat} is defined as the laser power P_L necessary to provide half of the maximum ion current I_{max} accessible

$$I_{\text{ion}}(P_L) = I_{\text{max}} \frac{P_L/P_{\text{sat}}}{1 + P_L/P_{\text{sat}}}. \quad (3.16)$$

However, for spectroscopy purposes saturation should be avoided since it introduces additional broadening [39, 40].

For simplification, assume a gas of identical non-interacting atoms with only two energy states: ground state E_g and an excited state E_e , which have the same statistical weights $g_g = g_e$. At time $t = 0$ all atoms are located in the ground state. If the gas is irradiated with monochromatic laser light of spectral energy density

$$\rho(\nu_0) = n(\nu_0) \cdot h\nu_0 \quad (3.17)$$

where $n(\nu)$ is the number of photons at transition frequency ν_0 , a portion of the photons will be absorbed and transfer electrons into the excited state. The population rate of the excited state can be expressed with the Einstein coefficients for stimulated transitions B and spontaneous emission A by

$$\frac{dN_e}{dt} = -AN_e + B\rho(\nu_0)N_g - B\rho(\nu_0)N_e, \quad (3.18)$$

where $N_{g,e}$ the atoms found in the ground and excited state. The three terms on the right-hand side in eq. (3.18) correspond to spontaneous emission, absorption, and stimulated emission, respectively. Since the total number of atoms $N_{\text{total}} = N_g + N_e$ is constant, eq. (3.18) can be simplified to

$$\frac{dN_e}{dt} = -AN_e + B\rho(\nu_0)(N_{\text{total}} - 2N_e). \quad (3.19)$$

If the gas is now irradiated with an intense light source such as laser radiation, stimulated emission dominates over spontaneous emission so that the first term in eq. (3.19) can be neglected. From this it can be seen that in the steady state condition where $\frac{dN_e}{dt} = 0$, the maximum number of atoms that can be found in the excited state is $N_e = \frac{1}{2}N_{\text{total}}$.

In reality, neither the probability of the atom to absorb a photon $g(\nu)$ nor the frequency of the laser are purely monochromatic; therefore, eq. (3.19) can be expressed more precisely as

$$\frac{dN_e}{dt} = -AN_e + B(N_{\text{total}} - 2N_e) \int_{-\infty}^{\infty} g(\nu)\rho(\nu)d\nu, \quad (3.20)$$

In order to determine the saturation-broadened linewidth of the transition, it is assumed that a laser with a much smaller linewidth scans across the resonance. In this case the spectral density can be described with the Dirac δ -function as $\rho(\nu) = \rho_0\delta(\nu - \nu_L)$, where ν_L is the laser frequency and ρ_0 is the energy of the laser per unit volume. If the laser is at resonance $\nu_L = \nu_0$, the number of atoms in steady state condition in an excited state can be written as

$$N_e^{\text{res}} = N_{\text{total}} \frac{B\rho_0g(\nu_0)}{A + 2B\rho_0g(\nu_0)}. \quad (3.21)$$

The linewidth of a transition can be defined as full-width half-maximum (FWHM) so that at a frequency ν_{FWHM} , the number of atoms in the excited state decreases by a factor of 50 %:

$$N_e^{\text{FWHM}} = \frac{1}{2}N_e^{\text{res}} = N_{\text{total}} \frac{B\rho_0g(\nu_{\text{FWHM}})}{A + 2B\rho_0g(\nu_{\text{FWHM}})}. \quad (3.22)$$

Substituting eq. (3.21) into eq. (3.22) gives

$$g(\nu_{\text{FWHM}}) = \frac{Ag(\nu_0)}{A + 2B\rho_0g(\nu_0)}. \quad (3.23)$$

Since $g(\nu)$ is normalized to $\int_{-\infty}^{\infty} g(\nu)d\nu = 1$, it can be seen that with increasing frequency ν the value for $g(\nu)$ must decrease. Therefore, it follows that with increasing laser intensity the transition

linewidth increases as well. In this case the transition's probability to absorb a photon is, due to Doppler broadening, given by a Gaussian

$$g(\nu) = \frac{1}{\sqrt{2\pi\Delta\nu_D^2}} e^{-\frac{(\nu-\nu_0)^2}{2\Delta\nu_D^2}} \quad (3.24)$$

with a linewidth $\Delta\nu_D$. Substituting eq. (3.24) into eq. (3.23), the transition linewidth at FWHM can be found from

$$\Delta\nu_{\text{Sat}} = 2|\nu_{\text{FWHM}} - \nu_0| = \Delta\nu_D \sqrt{8 \ln \left(2 + \frac{2}{\sqrt{2\pi}\Delta\nu_D} \frac{B}{A} \rho_0 \right)}. \quad (3.25)$$

Hence, for a Gaussian absorption probability in the steady state approximation, the saturation broadening increases with the square root of the natural logarithm of the laser intensity. In addition it can be noted that the ratio between the Einstein coefficients is $\frac{B}{A} = \frac{c^3}{8\pi h \nu_0^3}$ and thus, the linewidth decreases with increasing frequency. Since in resonant laser ionization excited states are coupled to the ionization continuum or further excited states, the steady state condition is not valid for this kind of experiments. Therefore eq. (3.25) only gives the order of magnitude for the expected broadening. For multi-step systems the coupled rate-equation must be solved. A typical approach would be to use the density matrix formalism which has been demonstrated in [41, 42].

3.4 Electron-nucleus interaction

In general, the atomic spectrum can be explained by electrons moving in a Coulomb potential created by the nucleus. However, if one takes a closer look it can be observed that the nucleus has further influences on the atomic spectrum. These effects are typically on the order of a few hundred MHz and thus cannot be detected with the experimental methods used in this work. On the other hand, in particular for heavy elements, these effects can increase up to the order of 10 GHz becoming resolvable with in-source resonant laser ionization spectroscopy. Apart from the nuclear charge Ze that creates the Coulomb field, the atomic spectrum is also affected by:

- nuclear motion responsible for the mass effect in the IS,
- nuclear size responsible for the field shift in the IS, and
- nuclear spin I and magnetic moment μ responsible for the HFS.

How these properties contribute to the atomic spectrum is illustrated in Figure 3.4 and will be further outlined.

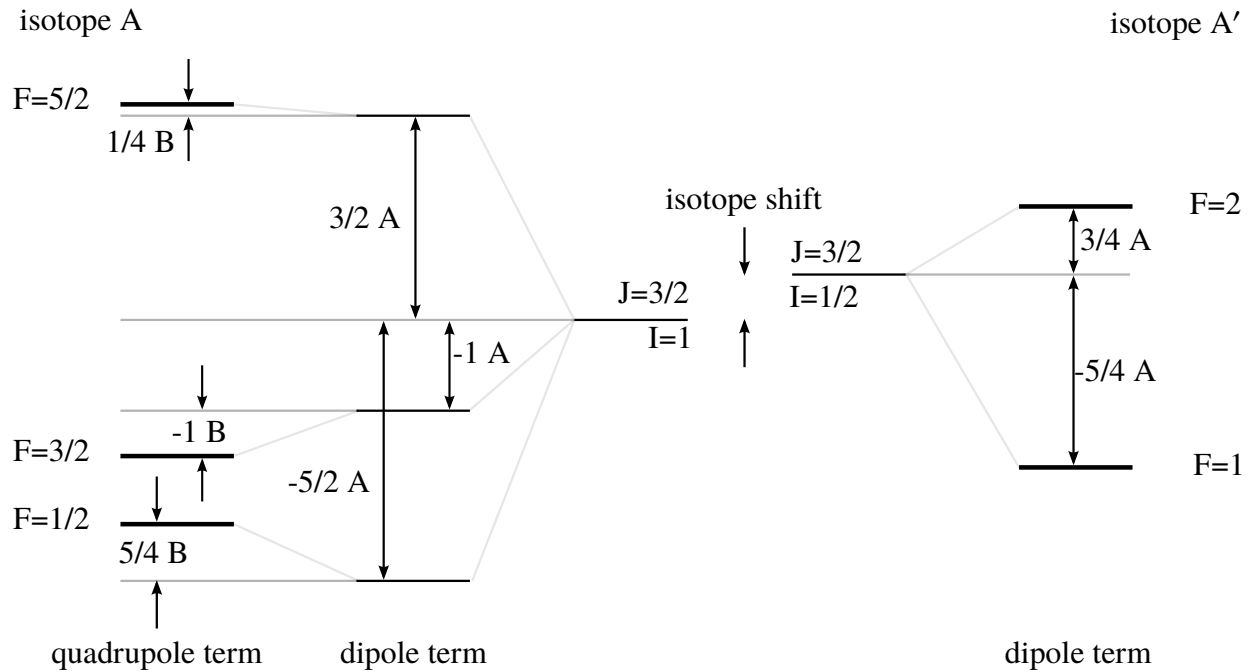


Figure 3.4: Hyperfine structure splitting and isotope shift of the same atomic state in two different isotopes A and A' of the same element.

3.4.1 Hyperfine structure

Protons and neutrons are spin $1/2$ -particles and therefore fermions. According to the nuclear shell model, these nucleons are arranged in shells similar to electrons in an atom [43]. Since the spins of paired nucleons cancel each other out, the total nuclear spin I results only from unpaired nucleons. Consequently, even-even nuclei have nuclear spin $I = 0$, odd-even and even-odd nuclei have half-integer spin, and odd-odd nuclei have integer spin.

Additionally, charged particles moving on an orbital path always create a magnetic moment μ which is, in the case of the nucleus, directly proportional to the nuclear spin:

$$\mu = g_I \mu_N I, \quad (3.26)$$

where g_I is the g -factor, a dimensionless proportionality constant between the spin and the magnetic moment. The unit of the magnetic moment is the nuclear magneton

$$\mu_N = \frac{e\hbar}{2m_p} = 3.152 \times 10^{-8} \text{ eV T}^{-1}, \quad (3.27)$$

where e is the elementary charge, and m_p is the proton mass. Thus, the nuclear magneton is a factor of 1836 smaller than the Bohr magneton describing the magnetic moment of an electron caused by its total angular momentum J . The magnetic field B_J created by the electrons penetrates into the nucleus and interacts the nuclear magnetic moment and the nuclear spin. As a result, the electron total angular momentum J and the nuclear spin I couples to a new total angular momentum

$$\hat{F} = \hat{I} + \hat{J} \quad (3.28)$$

similar to fine structure coupling with quantum numbers replaced as follows:

$$\begin{array}{ccc} \hat{L} & \hat{S} & \hat{J} \\ \downarrow & \downarrow & \downarrow \\ \hat{J} & \hat{I} & \hat{F}. \end{array} \quad (3.29)$$

For that reason, the fine structure energy level split into $2J + 1$ or $2I + 1$ (so called hyperfine structure levels) dependent on whether the electron total angular momentum J or nuclear spin I is larger. In addition to the selection rules in eqs. (3.2) to (3.4), optical transitions between the

hyperfine structure components are allowed if

$$\Delta F = 0 \pm 1 \quad \text{but} \quad F = 0 \nrightarrow F = 0, \quad (3.30)$$

is obeyed. Even if a transition is allowed, the intensities vary within the spectrum. The theoretical transition strengths are given by the so-called Racah intensities

$$S(F_g \rightarrow F_e) = \frac{(2F_e + 1)(2F_g + 1)}{2I + 1} \left\{ \begin{matrix} J_e & F_e & I \\ F_g & J_g & 1 \end{matrix} \right\}^2, \quad (3.31)$$

where $\{\dots\}$ is the Wigner $6j$ -symbol [44]. Since the magnetic moments are inversely proportional to the mass of the charged particle as shown in eq. (3.27), the hyperfine structure is approximately three orders of magnitude smaller than the fine structure and therefore typically only resolvable with high resolution spectroscopy techniques. In an observed spectrum, the position of a hyperfine structure component ν^{F_e, F_g} relative to the centre of gravity (CoG) of the transition is given by

$$\nu^{F_e, F_g} = \text{CoG} + \Delta\nu^{F_e} - \Delta\nu^{F_g}, \quad (3.32)$$

where F_e and F_g denote the total angular momenta of the excited and ground states, respectively. The energy splitting of each level is described by

$$\Delta\nu^F = A \frac{K}{2} + B \frac{\frac{3}{4}K(K+1) - I(I+1)J(J+1)}{2(2I-1)(2J-1)IJ}, \quad (3.33)$$

where K is

$$K = F(F+1) - I(I+1) - J(J+1), \quad (3.34)$$

and A and B are the hyperfine structure constants relating to the magnetic dipole moment, and electric quadrupole moment, respectively. Generally, the first term in eq. (3.33) is the main contribution to the hyperfine structure and is in most cases larger for the ground state than for the

excited state. The A -factor can be written as

$$A = \frac{\mu B_J(0)}{IJ}, \quad (3.35)$$

where μ is the nuclear magnetic moment and $B_J(0)$ is the magnetic field at the nucleus created by the valence electron. The latter is difficult to determine experimentally or by theoretical calculations, but in general it can be said that it is larger if the electrons are close to the nucleus i.e., for heavy nuclei, have small principle quantum number n and small angular momentum l [45]. It can be seen from eq. (3.35) that there is no hyperfine structure if either the nuclear spin $I = 0$ or the electron angular momentum $J = 0$ vanishes.

While the magnetic field at the nucleus is in general responsible for the splitting of the hyperfine structure, the electric field gradient $\left\langle \frac{\partial^2 V}{\partial z^2} \right\rangle$ produced from the shell electrons at the nucleus is responsible for small shifts in these energy levels. Up to this point it was assumed that the nucleus has a spherical symmetric shape. Most nuclei, however, have an ellipsoid shape resulting in an electric quadrupole moment Q . The electric quadrupole moment is defined as

$$Q = \frac{1}{e} \int (3z^2 - r^2) \rho(r) d^3r, \quad (3.36)$$

where e is the elementary charge, $\rho(r)$ is the charge distribution and, z is the direction of the axis of the rotational symmetry. It can be seen from eq. (3.36) that the Q -value for nuclei compressed along the z -axis is negative as shown in Figure 3.5; the shape of these nuclei is called oblate. Cigar-shaped nuclei stretched along the z -axis are called prolate and have a positive electric quadrupole moment. This is the most common shape of all existing nuclei [46].

It can be seen from eq. (3.33) that the second term vanishes if either the nuclear spin or the electron total angular momentum $I, J \leq 1/2$. This does not mean that these nuclei have spherical shapes, only that these deformations cannot be detected by evaluating the hyperfine structure [47]. The spectroscopic quadrupole moment Q_s differs therefore from the intrinsic quadrupole moment Q

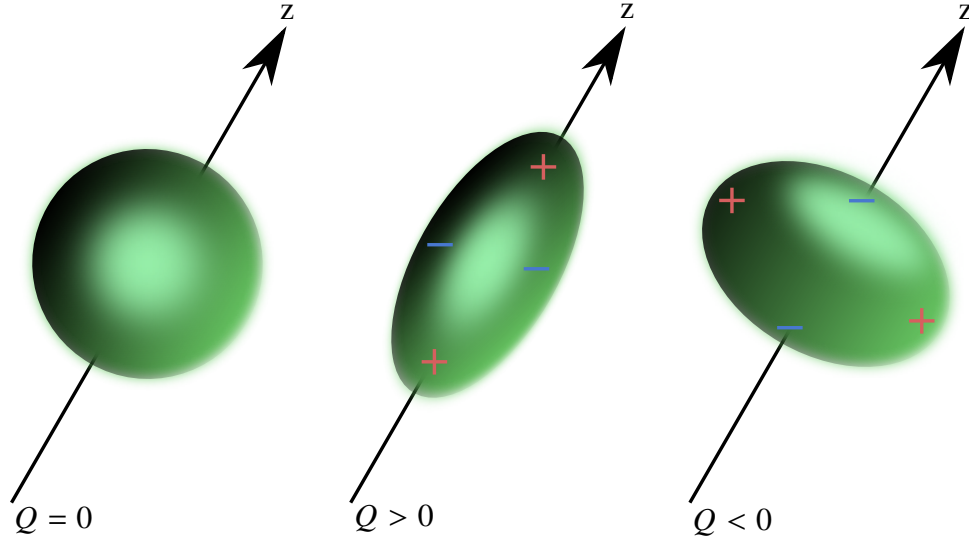


Figure 3.5: The charge distribution in an atomic nucleus is described in first order by the electric quadrupole moment and in general categorized as one of three different cases. For $Q = 0$ the charge is distributed spherically-symmetric. Where $Q > 0$, the nucleus has a cigar-shape (prolate). Finally, disk-shaped (oblate) nuclei have a negative quadrupole moment. The charge distribution is measured with respect to the z -component of the nuclear spin I .

and the hyperfine structure constant B becomes

$$B = eQ_s \left\langle \frac{\partial^2 V}{\partial z^2} \right\rangle. \quad (3.37)$$

For the spectroscopic quadrupole moment it is assumed that the rotational-symmetry axis is collinear with the nuclear spin I . If, however, a nucleus with prolate shape in the intrinsic frame rotates along an axis perpendicular to the symmetry axis, the observed time-averaged shape would be oblate [48]. For a well deformed nucleus the relation between the spectroscopic and the intrinsic quadrupole moments is

$$Q_s = \frac{3\Omega^2 - I(I+1)}{(I+1)(2I+3)}, \quad (3.38)$$

where Ω is the projection of nuclear spin along the deformation axis.

3.4.2 Optical isotope shift

Even though the transition energies of atomic spectra are element-specific, small differences in these energies can be observed between isotopes of atomic mass number A and A' . These energy changes are called isotope shift (IS) and can be defined as

$$\delta\nu^{A,A'} = \nu^{A'} - \nu^A. \quad (3.39)$$

The IS results from changes in the mass as well as the size of the atomic nucleus, and is therefore composed of two major terms the, mass shift $\delta\nu_{\text{MS}}^{A,A'}$ and the volume or field shift $\delta\nu_{\text{FS}}^{A,A'}$

$$\delta\nu^{A,A'} = \delta\nu_{\text{MS}}^{A,A'} + \delta\nu_{\text{FS}}^{A,A'}. \quad (3.40)$$

More detailed treatment of the optical IS can be found in literature [49, 50, 51].

Mass shift

The mass shift is correlated to the change in nuclear recoil energy occurring due to the difference in mass between isotopes A and A' . This shift can again be separated into two parts, the normal mass shift (NMS) and specific mass shift (SMS):

$$\delta\nu_{\text{MS}}^{A,A'} = \frac{m_A - m_{A'}}{m_A m_{A'}} (\text{NMS} + \text{SMS}), \quad (3.41)$$

where m_A and $m_{A'}$ are the nuclear masses of the corresponding isotopes. In the center-of-mass frame the atom itself is defined at rest; therefore, the nuclear momentum $\vec{P}_{\text{nuc}}$ can be replaced by the total momenta of the i th electron $-\sum_i \vec{p}_i$. The kinetic energy of the nuclear recoil motion for a

specific electronic state is thus defined as

$$\frac{\vec{P}_{\text{nuc}}^2}{2m_{\text{nuc}}} = \frac{(\sum_i \vec{p}_i)^2}{2m_{\text{nuc}}} = \frac{1}{2m_{\text{nuc}}} \left(\sum_i \vec{p}_i^2 + \sum_{i \neq j} \vec{p}_i \cdot \vec{p}_j \right), \quad (3.42)$$

where the $\vec{p}^2$ term relates to the NMS, and the $\vec{p}_i \cdot \vec{p}_j$ term to the SMS.

If an electron gets excited into a different state, the momenta of the electrons change and as a result, also their kinetic recoil energies. The NMS considers only the excited electron, thus the change in kinetic energy relates directly to the energy of the absorbed photon and the difference in the reduced masses of the electron-nucleus-system. It can be precisely calculated by

$$\text{NMS} = m_e \nu_0, \quad (3.43)$$

where m_e is the electron mass and ν_0 is the transition frequency of the reference isotope.

The NMS is the complete solution for a single electron atom as well as muonic atoms. For multi-electron atoms, correlations between electrons must be considered resulting in the specific mass shift. The SMS can only be determined numerically and to date was only accurately calculated for atoms with three electrons [52]. However, since the mass shift is proportional to $\delta\nu_{MS}^{A,A'} \propto 1/A^2$ it can, in first approximation, be neglected for heavy nuclei.

Field shift

The second term of the IS in eq. (3.40) is related to the electrostatic interaction between the electron and the nucleus. While outside the nucleus electrons experience a purely Coulomb potential $V(r) = -Ze/r$ equivalent for all isotopes of the same element, inside the nucleus the potential varies for different isotopes depending on their nuclear size as shown in Figure 3.6. Since s-electrons have the largest probability density inside the nucleus, these electrons are more affected

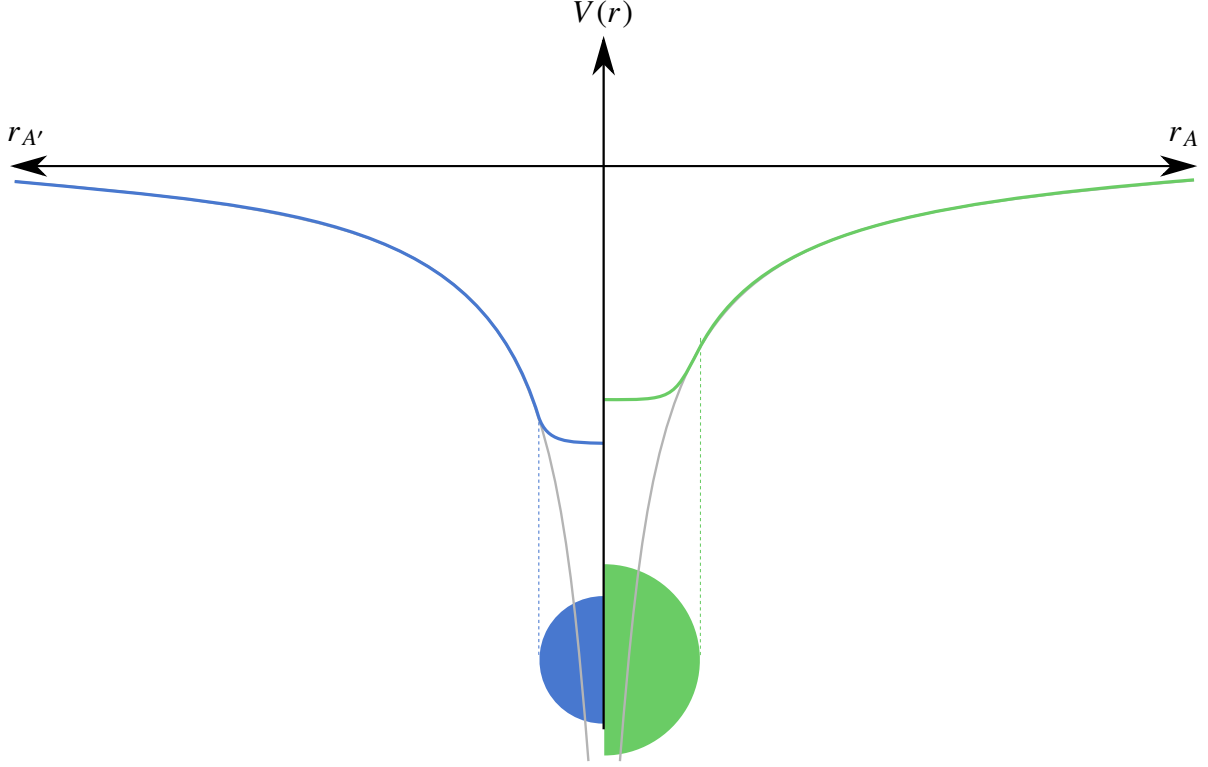


Figure 3.6: Modified Coulomb potential for two isotopes of different sizes.

by the potential difference. Therefore, the main contributions to the field shift

$$\delta \nu_{\text{FS}} = \frac{Ze^2}{6h\epsilon_0} \Delta |\Psi(0)|^2 \delta \langle r^2 \rangle = F \delta \langle r^2 \rangle \quad (3.44)$$

are the changes in electron density at the nucleus $\Delta |\Psi(0)|^2$ between the upper and lower states of the transition, and the nuclear mean squared charge radius $\delta \langle r^2 \rangle$. Since the atomic parameters stay constant for the same optical transition, they can be summarized to a field-shift constant F . The only nuclear variable is the mean squared charge radius defined as

$$\langle r^2 \rangle = \frac{\int 4\pi r^2 \rho(r) r^2 dr}{\int 4\pi r^2 \rho(r) dr}, \quad (3.45)$$

where $\rho(r)$ is the nuclear charge distribution. In the liquid drop model, $\rho(r)$ is assumed to be constant over the size of the nucleus so that eq. (3.45) can be simplified to

$$\langle r^2 \rangle = \frac{\int_0^R r^4 dr}{\int_0^R r^2 dr} = \frac{3}{5} R^2, \quad (3.46)$$

where the radius increases with increasing number of nucleons A as

$$R = R_0 A^{1/3}, \quad \text{with} \quad R_0 = 1.2 \text{ fm}. \quad (3.47)$$

Therefore, for small variations in A the mean squared charge radius changes by

$$\delta \langle r^2 \rangle = \frac{2}{5} R_0^2 A^{-1/3} \delta A. \quad (3.48)$$

Since the Coulomb potential as well as the electron density probability are each proportional to Z , it can be summarized that $\delta \nu_{FS} \propto Z^2/A^{1/3}$ [53] and the field shift is therefore the dominant term for heavy nuclei. The turning point where mass and field shift are approximately equal is at $Z \approx 38$ [50]. Equation (3.44) is only truly valid for light nuclei. For heavy isotopes higher radial momenta have to be included; thus, $\delta \langle r^2 \rangle$ needs to be replaced by the Seltzer moment $\lambda^{A,A'}$

$$\lambda^{A,A'} = \delta \langle r^2 \rangle + \frac{C_2}{C_1} \delta \langle r^4 \rangle + \frac{C_3}{C_1} \delta \langle r^6 \rangle + \dots, \quad (3.49)$$

where C_i are the Seltzer coefficients and are tabulated in [54]. So far it was assumed that the nucleus has a spherical shape, but as mentioned above the majority of nuclei have deformed shapes. For deformed nuclei the charge radius can, in first approximation, be written as

$$\langle r^2 \rangle \approx \langle r^2 \rangle_{\text{sph}} \left(1 + \frac{5}{4\pi} \langle \beta_2^2 \rangle \right) \quad \text{and} \quad (3.50)$$

$$\delta \langle r^2 \rangle \approx \delta \langle r^2 \rangle_{\text{sph}} + \frac{5}{4\pi} \langle r^2 \rangle_{\text{sph}} \delta \langle \beta_2^2 \rangle, \quad (3.51)$$

where $\langle r^2 \rangle_{\text{sph}}$ is the radius for a spherical nucleus with the same volume, and $\langle \beta_2^2 \rangle$ is the mean squared quadrupole deformation parameter.

King plot

The SMS as well as the field shift-constant F rely strongly on the input of theoretical calculation where accurate results can only possibly be obtained for the lightest atoms with up to three electrons. It is easy to imagine that those constants are not available for all possible transitions in every element. If, however, IS data is available for a transition in a muonic atom of an element of interest, x-ray spectra, or an optical transition where the SMS and F are known, it is possible to extract these for a different transition by the so-called King plot analysis [10, 49].

From the reflections above, the optical IS can be summarized to

$$\delta \nu^{A,A'} = \frac{m_A - m_{A'}}{m_A m_{A'}} (\text{NMS} + \text{SMS}) + F \lambda^{A,A'}, \quad (3.52)$$

where $\text{SMS} + \text{NMS} = M$ and F are transition-specific, and $m_A, m_{A'}$ and $\lambda_{A,A'}$ are isotope-specific parameters. In order to eliminate parameters related to $\delta \langle r^2 \rangle$, the IS for each transition i and j can be multiplied by a factor $\frac{m_A m_{A'}}{m_A - m_{A'}}$ resulting in a modified IS

$$\frac{m_A m_{A'}}{m_A - m_{A'}} \delta \nu_{i,j}^{A,A'} = M_{i,j} + F_{i,j} \frac{m_A m_{A'}}{m_A - m_{A'}} \lambda^{A,A'}. \quad (3.53)$$

Since $\lambda^{A,A'}$ is the only common factor, it can be substituted so that

$$\frac{m_A m_{A'}}{m_A - m_{A'}} \delta \nu_j^{A,A'} = \frac{F_j}{F_i} \left(\frac{m_A m_{A'}}{m_A - m_{A'}} \delta \nu_i^{A,A'} \right) + M_j - \frac{F_j}{F_i} M_i. \quad (3.54)$$

Therefore, a plot of the modified ISs of transition i against j results in a straight line with slope F_j/F_i , and intersection at $M_j - (F_j/F_i)M_i$. In addition to separating the transition specific parameters, the King plot analysis can also be used to examine the consistency of the measured ISs.

4 | Laser system

One of the requirements of laser systems used for laser ion source operation is a large tuning range in order to access resonances for as many elements as possible. The strongest transitions of the first excitation step (FES) for most elements are in the ultraviolet (UV) to blue region, while the second excitation step (SES) often lies in the visible spectrum (VIS) to near infrared (NIR). Commonly used for their wide tuning ranges are dye lasers, covering a wavelength range from the UV to NIR by switching between different dyes and pump lasers. These lasers are used at the one of the first laser ion sources in operation at ISOLDE-CERN [5, 55]. TRILIS at TRIUMF uses Ti:Sa lasers described in section 4.1, which lase in the NIR region. Shorter wavelengths from the UV to blue region can be reached by frequency conversion, further discussed in section 4.2. The missing gap, the VIS, may either be covered with a combined system of dye and Ti:Sa lasers [56, 57], difference frequency mixing demonstrated on the sodium D2 line at 589.158 nm [58], or partly (584 nm to 672 nm) with the frequency-doubled light of a Cr:Forsterite laser [59]. In addition to the Ti:Sa lasers, TRILIS operates a high-power neodymium-doped yttrium orthovanadate (Nd:YVO₄) laser (section 4.3) for higher efficiency of nonresonant ionization.

4.1 Titanium sapphire laser

Titanium-doped sapphire (Ti:Sa) crystals are the most widely used, tunable, solid state laser mediums. They consist of corundum crystals (Al₂O₃) in which 0.1 atomic percent of the Al³⁺ are

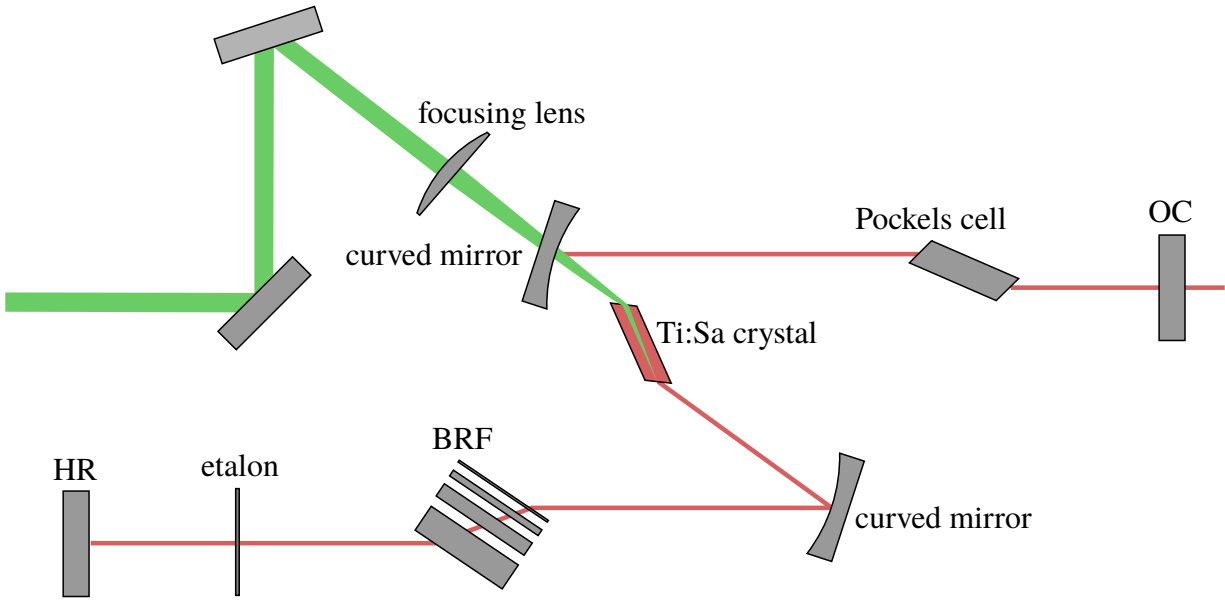


Figure 4.1: BRF Laser

replaced by Ti^{3+} ions, which are responsible for the characteristic wide emission spectrum from 600 nm to 1100 nm [60]. The emission peak is at 780 nm with a FWHM of about 180 nm [61]. The absorption spectrum is between 400 nm to 650 nm with a peak at 488 nm [60]. Due to an overlap of the emission and absorption spectra and their wide tails, the tuning range in laser operation is between 680 nm to 1000 nm.

A pump source for the Ti:Sa lasers at TRIUMF is realized by a frequency-doubled Nd:YAG laser¹ emitting a wavelength of 532 nm. It provides 50 W of laser power which can be divided between the maximum four lasers on the table.

The resonator design of TRILIS Ti:Sa lasers, as shown in Figure 4.1, is based on a Ti:Sa laser developed at the Johannes Gutenberg-University (Mainz, Germany) [62], which is optimized for operating conditions at on-line, ISOL type, radioactive ion beam facilities (i.e. pulsed operation to make use of the high pulse peak power, at repetition rates exceeding 10 kHz). It has a linear, Z-shaped resonator, with two resonator mirrors of curvature $R = 75$ mm in whose center the Ti:Sa laser is located, and two mirrors at the end of parallel arms orientated at 36.2° to the resonator

¹LEE LPD 100 MQG

arm. This angle is chosen to compensate for the astigmatism introduced by the Brewster angles [63] on the Ti:Sa crystal and reduces reflection losses on its surfaces. In the original design, the focus was set to the front of the crystal whereas in the new designs two different crystal positions can be chosen from. In order to better match the pump mode with the resonator mode, the focus is set to the far crystal surface. The advantage of having the focus on the back side is that the laser can be pumped harder without damaging the crystal.

All mirrors except the output coupler have a reflectivity near 100% at their center wavelength, whereas the resonator mirrors have an additional anti-reflection coating for 532 nm in order to minimize the pump beam transmission losses. Since the TRILIS laser system runs in pulsed operation with a repetition rate of 10 kHz, the output coupler has a reflectivity of 70 % to 80 %.

The advantage of pulsed laser systems is the availability of high peak intensities and linewidths in the 0.6 GHz to 7 GHz region. High laser intensities are required to saturate weak excitation steps and for frequency conversion in nonlinear crystals needed to reach wavelengths in the blue and UV frequency region. The typical average power the TRILIS Ti:Sa lasers provide at 10 W pump power is between 1 W to 3 W at a pulse length of about 40 ns [64].

Since for resonant ionization the laser pulses have to reach the ionization region simultaneously, each laser can be equipped with a Pockels cell. The Pockels cell is based on the electro-optical effect, and consists of a nonlinear crystal potassium dihydrogen phosphate (KDP) which converts linear polarized laser light to circular polarized under application of a voltage of 1.5 kV to 6 kV. Due to the change of polarization, additional losses are introduced associated with the resonator which spoils the resonator Q-factor. By turning off the voltage, the crystal becomes transparent and the conversion can be accessed again [65]. If the time lag between two pulses is short, the synchronization can be realized passively by either adjusting the pump power, or changing the Ti:Sa crystal position in one of the resonators. The closer the focus is located to the front crystal surface, the earlier the laser starts lasing.

For laser ion source operation, a laser linewidth which best fits the Doppler broadened spectrum is desirable in order to excite atoms of all velocity classes. Typical Doppler broadened linewidths for a source temperature of 2000 °C are between 1 GHz to 4 GHz. If the laser is used to scan over a resonance in order to look for possible hyperfine structure, a narrow laser linewidth is desirable, since the resulting measured linewidth $\Delta\nu_R$ is always a convolution of the laser linewidth $\Delta\nu_L$ and natural atomic linewidth $\Delta\nu_A$. For a Gaussian lineshape or near-Gaussian, this can be calculated exactly

$$\Delta\nu_R = \sqrt{\Delta\nu_L^2 + \Delta\nu_A^2}. \quad (4.1)$$

However, the linewidth of a plain resonator is typically several THz wide, resulting from the emission spectrum of the gain medium and the reflection curves of the resonator mirrors. In order to reach the required linewidth, additional wavelength-selective elements are placed in the resonator.

The common method for wavelength selection in the TRILIS Ti:Sa-laser is the combined use of a birefringent filter (BRF) and an etalon. The features of this laser are its long time frequency stability, narrow linewidths, and high laser intensities of 1 W to 3 W average power at 10 W pump power.

4.1.1 Birefringent Filter

The BRFs² mostly used in TRILIS lasers consist of four birefringent crystal plates with thickness ratios of 1:4:8:15 and a base thickness of 0.5 mm. Linear polarized light passing through the birefringent plate is divided into an ordinary and an extraordinary beam, travelling in the material with different velocities $v_o = c/n_o$ for the ordinary beam and $v_{eo} = c/n_{eo}$ for the extraordinary beam. Both beams are polarized perpendicular to each other. If the two beams have a phase difference

$$\delta \neq 2m\pi \quad \text{with} \quad m = 1, 2, 3 \dots \quad (4.2)$$

²VLOC BFU38.1-1T4T8T15T.

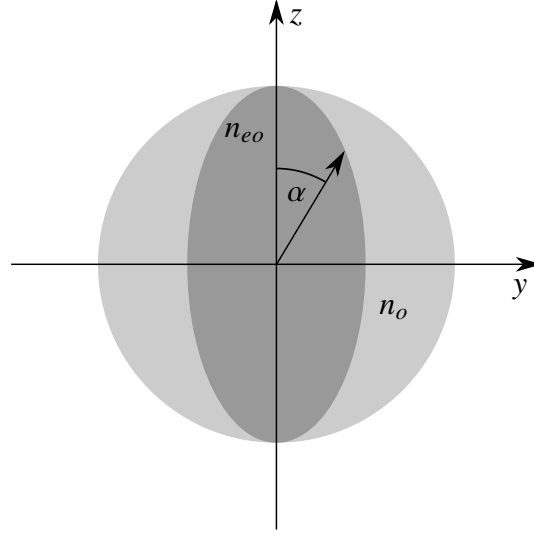


Figure 4.2: Index ellipsoid for birefringent materials. The extraordinary index of refraction is dependent on the angle α between the direction of propagation and the optical axis.

after passing through the plate, the resulting beam is elliptically polarized. All surfaces in the resonator which are not mirrors are chosen as Brewster surfaces in order to reduce reflection losses, therefore beams which are not p-polarized are suppressed. Wavelengths λ passing through the filter

$$\lambda = \frac{\Delta n \cdot d}{m} \quad \text{with} \quad m = 1, 2, 3 \dots \quad (4.3)$$

therefore depend on the difference between the indexes of refraction $\Delta n = n_o - n_{eo}$ and the thickness d of the thinnest plate. Since the extraordinary index of refraction n_{eo} depends on the angle α between the direction of propagation and the optical axis shown in Figure 4.2, the wavelength can be selected by rotating the optical axis of the crystal around the direction of propagation of the light. As the resulting transmission curve shown in Figure 4.3, is the product of the transmission curve of each plate, the thinnest plate is responsible for the free spectral range (FSR) of the filter, i.e., the spectral distance between two transmission peaks. The thickest plate is crucial for the laser linewidth.

In order to prevent the laser from running on two wavelengths simultaneously, a large FSR must be chosen. However, as shown in eq. (4.3), the condition for transmission is valid for several

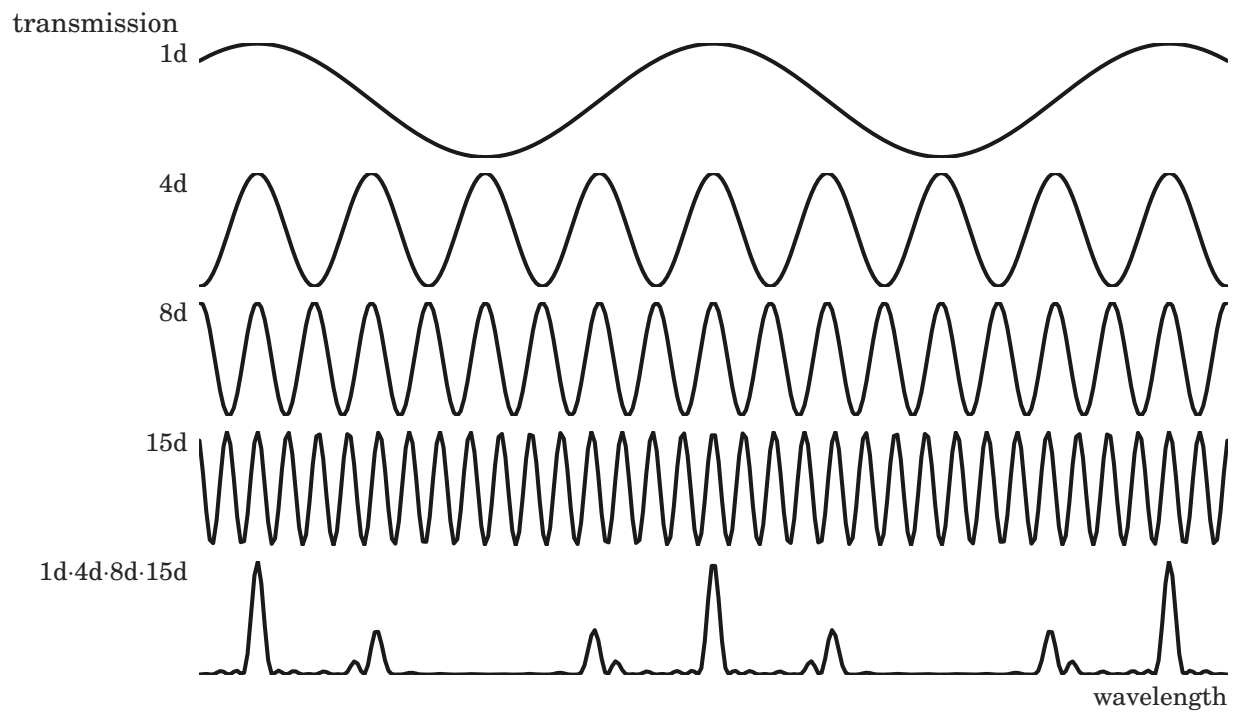


Figure 4.3: Transmission curves of the TRILIS four plate birefringent filter where the plates have a thickness ratio of 1:4:8:15. The final transmission (bottom panel) is the product of the individual plate transmission curves (upper panels). The free spectral range is defined by the thinnest plate, and the linewidth by the thickest.

wavelengths simultaneously. For that reason it is important, particularly if wavelengths beyond the emission maximum are required, to choose mirrors which suppress lasing in the maximum. The further away from maximum the chosen wavelength is, the narrower the reflectivity required for the mirror. The laser linewidth reachable with BRF has been measured to be $\Delta\nu \approx 100$ GHz [62] which is, for most cases, too wide for efficient excitation. In order to narrow the linewidth further, an etalon can be added.

4.1.2 Fabry-Pérot etalon

A solid Fabry-Pérot etalon is basically a thin glass plate of thickness d and partly reflecting coatings with reflectivity R on each surface. Due to interference effects, only wavelengths which are integer multiples of its thickness can be transmitted. In order to change the wavelength, the etalon can be tilted by an angle β which increases the path length light travels in the material. Since the linewidth provided by the BRF is $\Delta\nu \approx 100$ GHz, the FSR of the etalon should be larger in order to prevent the laser from running in multiple spectral modes. Similar to the FSR of the BRF, the etalon's FSR also depends on its optical thickness:

$$\text{FSR} = \frac{c}{2nd} . \quad (4.4)$$

It follows that an etalon, consisting of fused silica ($n = 1.45$), should have a maximum thickness of

$$d \lesssim \frac{c}{2 \cdot 1.45 \cdot 100 \text{ GHz}} = 1 \text{ mm} . \quad (4.5)$$

The linewidth of the light transmitted depends on the number of partial beams it can interfere with. This number is described by the Finesse F of the etalon, regulated by its reflectivity R as

$$F = \frac{\pi\sqrt{R}}{1 - R} . \quad (4.6)$$

The higher the reflectivity, the larger the finesse. The actual linewidth is given by the ratio of the FSR to the finesse

$$\Delta\nu = \frac{\text{FSR}}{F} . \quad (4.7)$$

With these laser linewidths $\Delta\nu < 1$ GHz can be reached; however, it typically operates at about 3 GHz. This linewidth is in a comfortable range, matching well with the Doppler-limited absorption profile of the atoms in the hot ionization region of the on-line target ion source.

4.2 Harmonic frequency generation

Since the strongest first excitation steps of atoms are typically in the blue to UV spectrum, a frequency conversion of the NIR wavelength emitted from the Ti:Sa-lasers is necessary. This frequency conversion is realized by nonlinear polarization effects in optical nonlinear crystals [40]. In this way, two monochromatic waves at frequencies ν_1 and ν_2 can be converted into a third wave with frequency $\nu_3 = \nu_1 + \nu_2$. Typical crystal materials used at TRILIS are β -barium-borate (BBO) known for its high damage threshold [40], bismuth-borate (BiBO) for its high conversion efficiency, and lithium-triborate (LBO) for its good beam profile due to small walk-off angle³⁴. The phase matching for these crystals is realized by adjusting the tilting angle between the incoming laser beam and crystal surface.

The easiest case of frequency conversion is frequency-doubling in which two photons of the same beam can be converted into one photon with twice their energy. In this way, wavelengths between 345 nm to 495 nm are reachable. By frequency-tripling, a combination of the doubled and the fundamental light; and frequency-quadrupling, where the fundamental is doubled twice, the wavelength spectrum can be extended from 330 nm to 210 nm. A schematic drawing of the TRILIS tripling unit is shown in Figure 4.4.

³as specified by the manufacturer www.crystech.com.

⁴separation between ordinary and extraordinary waves in nonlinear crystals.

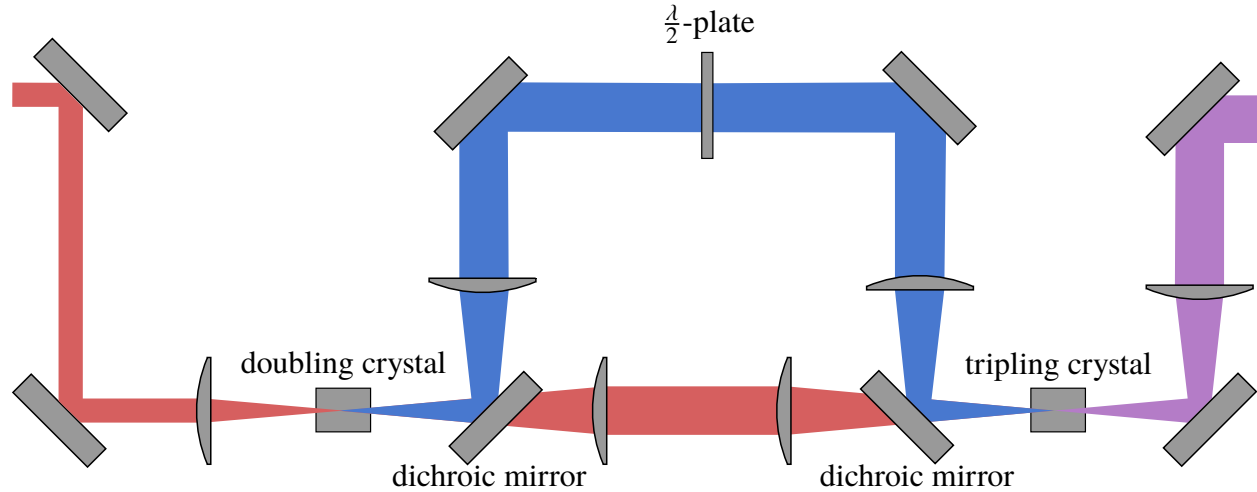


Figure 4.4: Schematic drawing of a frequency tripling unit. The NIR fundamental light of the laser is focused on a angle tuned doubling crystal. The resulting blue and NIR light is separated by a dichroic mirror in order adjust the polarization of the blue light. Finally the both laser beams are again combined and overlapped in the angle tuned tripling crystal.

The doubling efficiency of a nonlinear crystal is proportional to the squared electric field intensity E^2 . Since TRILIS uses pulsed lasers, the peak intensity of a single pulse is sufficiently high that the frequency can be converted by only one pass through the crystal. For that reason, it is sufficient to focus the laser on the crystal surface. A good beam profile (transverse electromagnetic mode $(\text{TEM})_{00}$) [63] is mandatory. Depending on which nonlinear crystal is used, the beam profile of the converted light may have an elliptic shape, which can be compensated for by introducing a set of cylindrical lenses.

Another possibility for frequency-doubling is to place the crystal inside the laser cavity. Due to the high intra cavity intensity, no focusing is necessary, which provides a similar output with a wider beam inside the crystal. For intra cavity doubling, the output coupler can be replaced with a high reflector for the fundamental, that is also highly transparent to the doubled light. Since the crystal emits light in both directions in the resonator, the blue output power can be increased by placing a dielectric mirror behind the crystal. The advantages of intra cavity doubling compared to external doubling are therefore an easy setup, high output powers, better beam profiles, and longer crystal lifetimes.

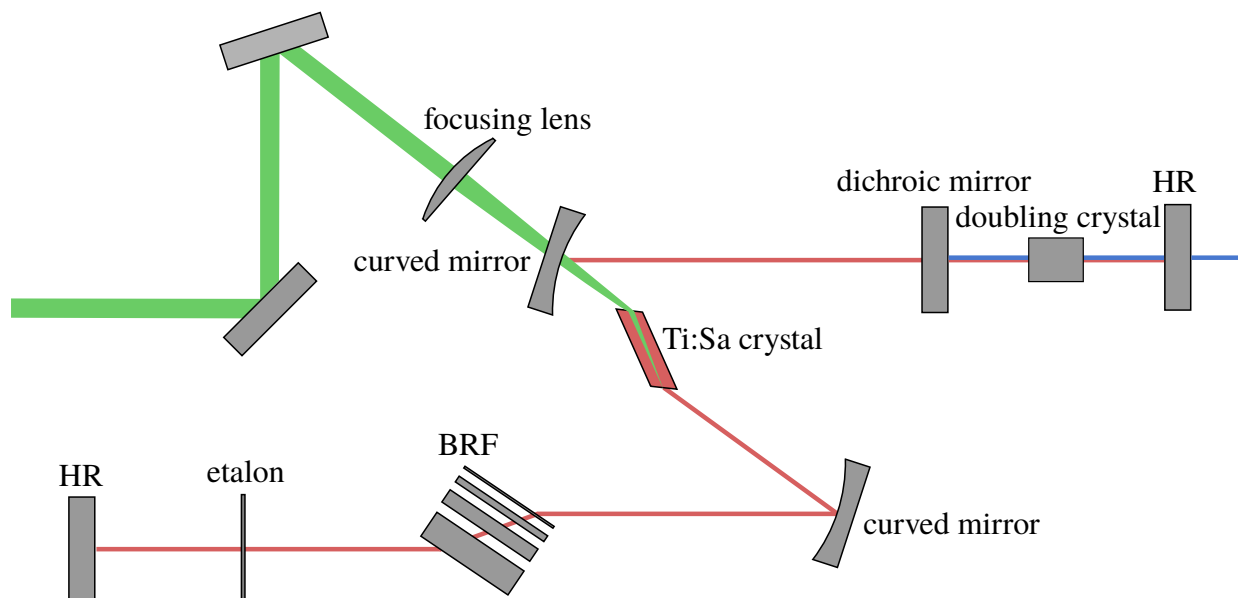


Figure 4.5: Ti:Sa laser cavity with intracavity doubling.

4.3 Ionizing laser

As shown in section 3.2.1, the laser power necessary to saturate a nonresonant transition is about 9 W, which cannot be provided by the common Ti:Sa lasers. For many elements no efficient resonant ionization step is known, so nonresonant ionization is the most common and reliable method of ionization. To address this, a diode-pumped Nd:YVO₄ laser⁵ completes the TRILIS laser system. In its fundamental it provides 13 W at 1064 nm, a frequency-doubling unit is available providing 6.7 W at 532 nm. Since in most cases the energy difference from the SES to the IP is considerably large, the frequency-doubled Nd:YVO₄ laser is the used most often. The advantages of using the nonresonant ionization laser instead of the pump laser are that it can be individually synchronized with the Ti:Sa lasers, it has good beam quality of TEM₀₀, $M^2 < 1.2$ [66], which leads to higher intensities at the focus, it has a similar pulse length as Ti:Sa lasers, and can be synchronized externally.

⁵Spectra-Physics YHP40 Navigator II.

5 | Silver

From the laser ion source point of view, silver is one of the most interesting elements: its atomic ground state can reach HFS splittings on the order of $\Delta\nu = 40$ GHz. Nearly every silver isotope has in addition to the nuclear ground state, an isomeric state with half-lives from $t_{1/2} = 1.5$ ms to 418 y. Since the nuclear spin of those two states can be very different, it is possible to separate the nuclei in the ground state from those in an isomeric state. This additional RIB purification is of interest for many experiments.

Since the yields for ground and isomeric states can differ by some orders of magnitude, simple ion detection on a CEM or FC is insufficient for observing the different resonances belonging to those two nuclear states. In this case it is of interest to take advantage of the different decay properties that can be monitored at the ISAC Yield Station as described in section 2.4.3. The main purpose of this chapter is the introduction of radioactive decay detection combined with resonance ionization spectroscopy (RADRIS), first reported in 1992 by W. Lauth [67].

Silver isotopes at ISAC are requested for the purpose of studying both the slow and rapid neutron capture processes (s-process and r-process). The s-process is a process that occurs inside of star at relatively low neutron-fluxes and temperatures. In order to study the stellar abundances of rare isotopes in the Cd-In-Sn region, an isotope beam of ^{115}Ag is requested, decaying by β -emission into an excited state of ^{115}Cd whose γ -spectrum was studied in experiment S1007¹. The presented data for $^{114,115}\text{Ag}$ in section 5.2 was taken in preparation for delivering RIB to this experiment.

¹Equilibrium of $^{115}\text{Cd}^m$ during the s-process, available at: <https://mis.triumf.ca/science/experiment/view/S1007>.

The r-process compared to the s-process, occurs at, high neutron-fluxes and temperatures that can be reached during supernovae. Studying the nuclei of $^{128-130}\text{Ag}$ is of interest, since they are located around the neutron shell closure at $N = 82$. Experiment S1542² was scheduled to investigate the γ -spectrum and lifetime of these isotopes, but as a consequence of a broken target module, it was not possible to deliver the requested ion beam purity and intensity.

The expected yields for these neutron-rich silver isotopes are on the order of 10 cps to 400 cps, but only if efficiently laser-ionized. Resonant laser ionization, is one method that can be used to produce the required isotopes with high efficiency, if the atomic excitation frequencies are known. The HFS and/or IS were only studied for neutron-deficient silver isotopes $^{97-110}\text{Ag}$ [68, 69] and select neutron-rich isotopes $^{122-127}\text{Ag}$ [70]. In order to get a better understanding of where the resonances for the requested isotopes are expected, the heaviest isotopes $^{116-119}\text{Ag}$ that could still be extracted from the broken target were studied. The results are shown in section 5.3.

5.1 Laser setup for silver resonance ionization

Silver was ionized via a three-step excitation scheme [71] (Figure 5.1) where the ionizing step was realized with the nonresonant frequency-doubled ionization laser at 532 nm. Although silver is one of the most interesting elements for resonant laser ionization due to the possibility of isomer separation, it is also the most difficult to implement with Ti:Sa laser systems. The FES can only be achieved by frequency tripling a fundamental wavelength of 984.489 nm, which is at the edge of the Ti:Sa crystal emission spectrum. Since a BRF-filter introduces only small losses in the resonator, the laser is forced to lase at the required wavelength by using a mirror set specialized for this wavelength region. Therefore, a high reflector was chosen with a center wavelength of 1010 nm and a cut off wavelength of 894 nm. The output coupler was originally used for the fundamental

²Investigation of lifetimes in neutron-rich Ag nuclei and structure on their Cd decay products, available at: <https://mis.triumf.ca/science/experiment/view/S1542LOI>.

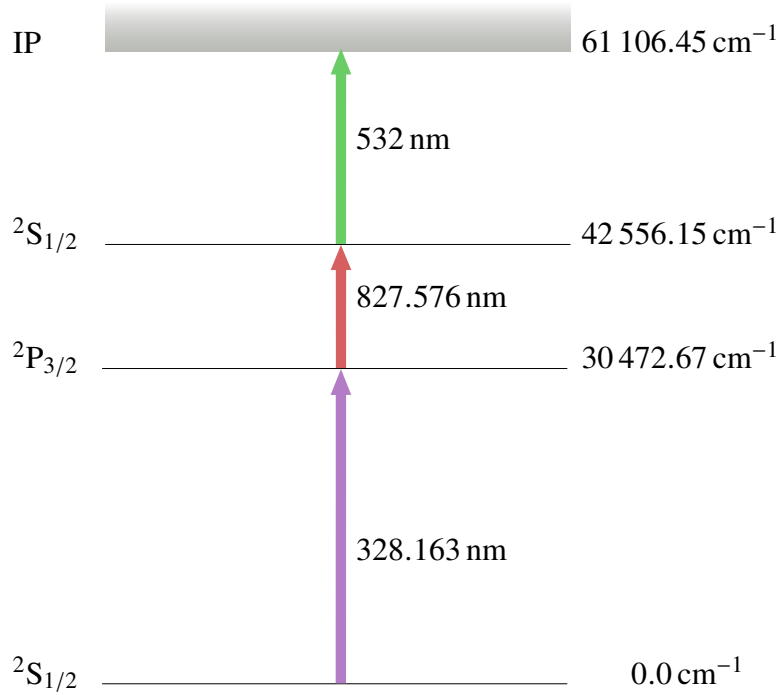


Figure 5.1: Silver excitation scheme using a two-step resonant excitation followed by nonresonant ionization.

wavelength of a 1064 nm Nd:YAG laser; at the requested wavelength, this mirror has a reflectivity of 70 %.

5.2 β decay evaluation

The data presented in this section was collected in preparation to deliver ^{114,115}Ag RIB to an experiment. The silver isotopes were produced by impinging a tantalum target with a 60 μ A, 480 MeV proton beam. The target was connected to a standard surface ion source. Prior to every beam delivery, the RIB has to be characterized; i.e. the yield of the requested isotope including any possible isobaric contamination needs to be determined. In the case of ¹¹⁴Ag and ¹¹⁵Ag, the isobaric background mainly consists of the surface-ionized stable ^{115g}In, and unstable ^{115m}In, ^{114g,m}In, having half-lives of $t_{1/2} = 4.5$ h, $t_{1/2} = 71.9$ s, and $t_{1/2} = 45.5$ d, respectively.

The electron transition frequencies for the ground and isomeric nuclear states would commonly be optimized by scanning the laser frequency manually and monitoring the change of the ion current on a CEM or FC signal. In this case, the isobaric background of the stable or close-to-stability indium isotopes was overwhelming and did not allow for an observation of a change in the overall ion current when the lasers needed to ionize silver were introduced. Even though it is possible to set up the laser for the FES at the requested wavelength, continuous scanning without emitting an additional longitudinal mode is challenging. For that reason, in order to find the correct frequencies, the laser wavelength was manually changed by tilting the etalon, and in case of any unwanted longitudinal mode appearing in the interference pattern at the wavelength meter³ the BRF filter was readjusted. At every wavelength a new yield measurement was performed and the yield was determined by evaluating the β decay of the produced RIB.

The ISAC target emits under ideal conditions a constant beam of N unstable nuclei per second, leading to a constant release of activity $A(t)$

$$A(t) = \lambda N(t), \quad (5.1)$$

where $\lambda = \frac{\ln 2}{t_{1/2}}$ is the decay constant. This activity is used to determine the yield of the RIB. For that purpose the activity is collected on a tape in the ISAC Yield Station for a time interval $t_{collect}$, the collection time. The implanted nuclei $N(t)$ constantly decay as described by an exponential decay function

$$N(t) = N_0 \cdot e^{-\lambda t}. \quad (5.2)$$

Beginning the implantation at $t = 0$ on a clean tape, the activation rate P of the tape can be expressed as

$$P = \frac{\lambda N(t_{collect})}{1 - e^{-\lambda t_{collect}}}. \quad (5.3)$$

³High Finesse WS7

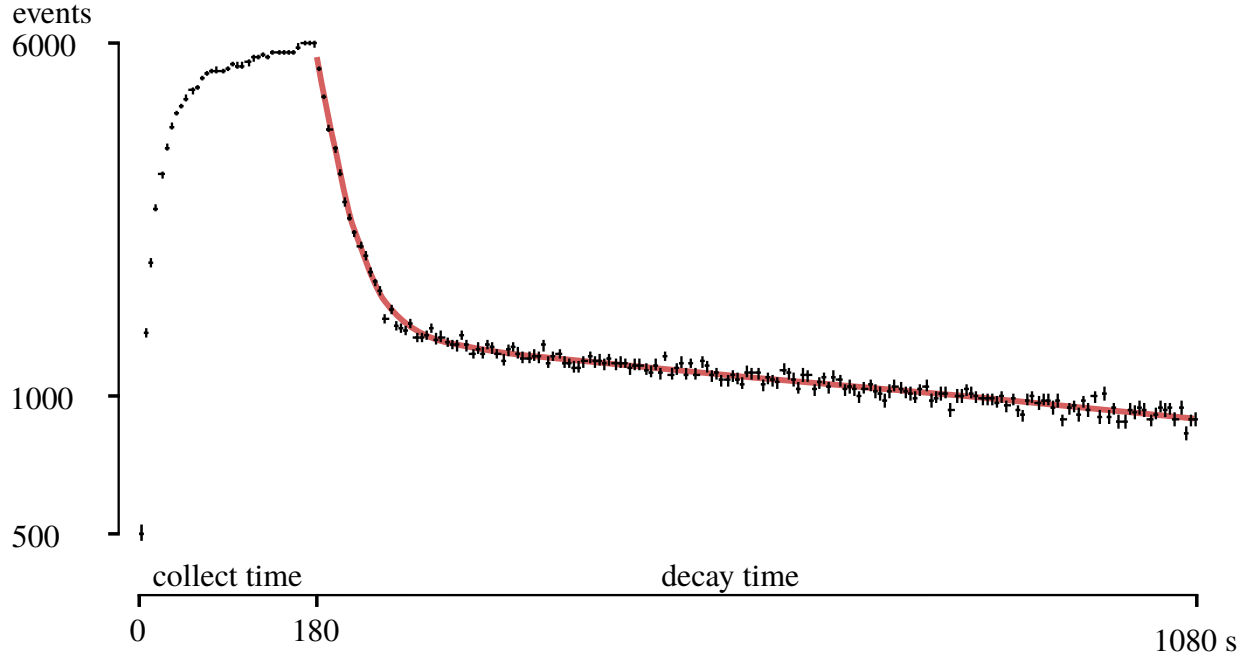


Figure 5.2: β decay for ^{115}Ag , with measurement intervals of $t_{\text{collect}} = 180$ s, $t_{\text{decay}} = 900$ s. The decay components of the short-lived isomer and the long-lived ground state can be clearly identified.

In order to determine the number of implanted nuclei $N(t_{\text{collect}})$, the RIB is blocked in the mass separator area for a decay time t_{decay} , and the α , β , and γ particle emitted from the radioactive isotope are monitored with the appropriate detector (see section 2.4.3); for β -radiation, the detectors are a set of scintillators coupled to photomultiplier tubes. Subsequently, the part of the tape with implanted activity can be moved behind a lead shielding, and the process repeated if desired to increase statistics.

For β radiation, an exponential decay-function eq. (5.2) is fitted to the data, where $N_0 = N(t_{\text{collect}})$. Such a β decay curve for ^{115}Ag with a laser set at a wavelength at which both components are ionized: the long-lived ($t_{1/2} = 20$ min) ground and the short-lived ($t_{1/2} = 18$ s) isomeric state, is shown in Figure 5.2. In order to convert the activation rate into the actual yield, the number of measurement cycles n , and the detector efficiency ϵ have to be taken into account. The yield of a

specific isotope can then be expressed as

$$\text{yield} = \frac{P}{n\epsilon}. \quad (5.4)$$

This yield measurement was repeated with different wavelengths of the FES over a range of ≈ 80 GHz. The measurement settings for each isotope are listed in Table 5.1. In order to fit the

Table 5.1: Yield measurement settings for $^{114,115}\text{Ag}$

isotope	$t_{1/2}$ [s]	number of cycles	t_{collect} [s]	t_{decay} [s]
^{114g}Ag	4.6	3	1	10
^{115g}Ag	1200	1	180	900
^{115m}Ag	18	2	20	80

decay-function to the β -decay curve, the half-life was fixed to the literature values take from [72]. Even though ^{114}Ag also has an isomer [73], it cannot be extracted from the target since its half-life, $t_{1/2} = 1.5$ ms, is too short for a significant number of atoms to reach the ionization region before they decay.

The results from the wavelength-dependent yield measurements are shown in Figure 5.3. Since Doppler broadening is the dominant broadening mechanism, each spectrum was fitted with up to two Gaussian peaks, depending on the magnitude of HFS splitting. In order to fit the data, a χ^2 minimization routine was used. At first the data was fitted without weights to determine the background. Afterwards the background was subtracted for the measured yields, and the uncertainty of the previous fitted background σ_{BG} was added to the existing yield error σ_{raw}

$$\sigma_y = \sigma_{\text{BG}} + \sigma_{\text{raw}}. \quad (5.5)$$

Typical uncertainties of yield measurements are on the order of 10 %. In addition, a constant wavelength error of $\sigma_x = 0.5$ GHz was included. This uncertainty was extracted from the wavelength fluctuations monitored over one week of beryllium RIB delivery, and it corresponds well with the precision of 0.001 nm for lasers with GHz linewidth quoted in the wavelength meter manual [74].

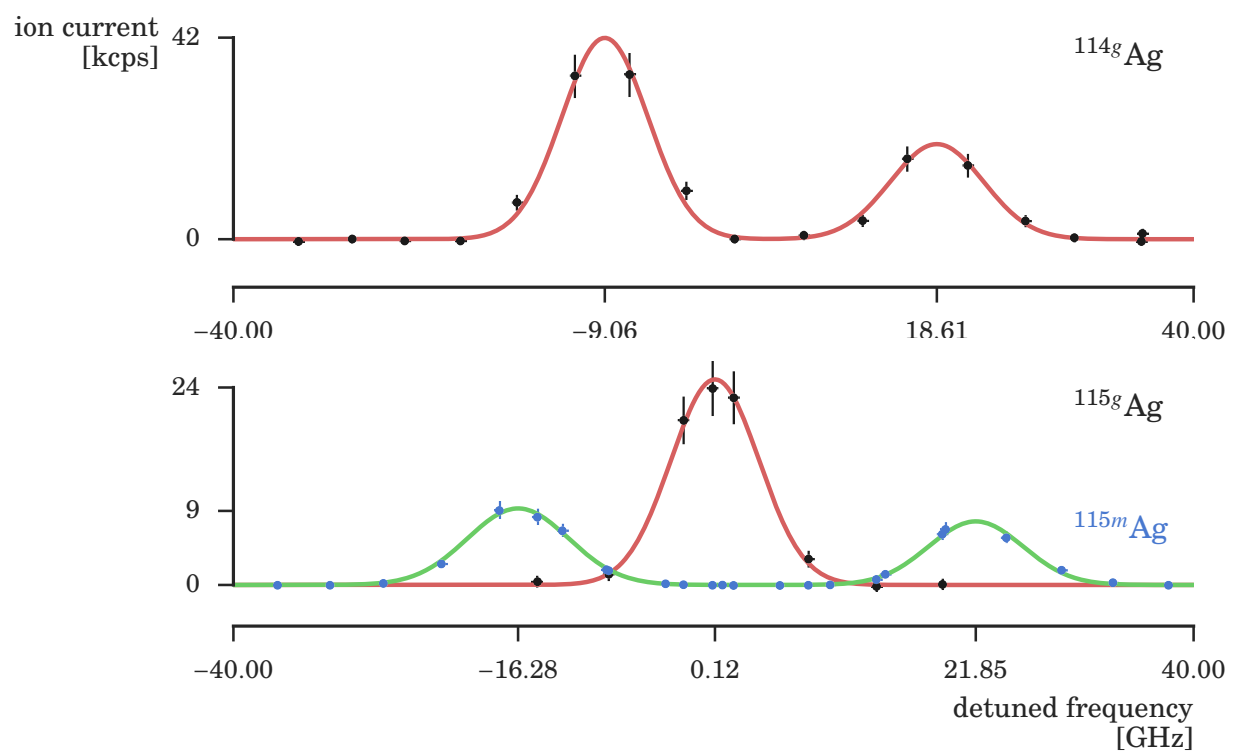


Figure 5.3: Frequency scan of the first excitation in $^{114,115}\text{Ag}$. The scan frequency is relative to $30\,472.67\text{ cm}^{-1}$.

The uncertainty for each data point can then be calculated to

$$\sigma_i^2 = \sigma_y^2 + \left(\frac{\partial f(x)}{\partial x} \right)^2 \sigma_x^2, \quad (5.6)$$

where $f(x)$ is the fit function. It should be noted that this assumption is only truly valid for linear equations with Gaussian error distribution [75].

5.3 γ decay evaluation

Cesium has its production maximum around mass $A = 130$, and a surface ionization efficiency of almost 100 %. The experiment to study the γ spectrum of $^{128-130}\text{Ag}$ is therefore only feasible if the isobaric cesium isotopes can be suppressed. For that reason, this experiment was planned to run with a UC_x -target in combination with an IG-LIS target module. However, the repeller electrode on this particular target module had shorted to the source high voltage so that both the isobaric suppression as well as the isotope release were hindered. It was therefore not possible to deliver the requested isotopes. As the yield for $^{128-130}\text{Ag}$ is expected to be low and the scheduled experiment was not able to collect a meaningful data with the extractable isotopes, this time was allocated for frequency scans of the FES in the heaviest extractable silver isotopes $^{116-119}\text{Ag}$. With these data it may be possible to interpolate the IS, and therefore obtain information on the expected resonances for $^{128-130}\text{Ag}$.

For continuous scanning of the laser frequency for the FES, a second etalon was placed inside the resonator cavity. This additional etalon not only reduced the laser frequency linewidth, but also introduced additional losses inside the laser cavity so that possibly appearing longitudinal modes can be suppressed. In order for the laser frequency to be scanned remotely with the ISAC Yield Station program, an etalon inside the laser cavity was mounted in a motorized mirror mount. When the yield program is set for laser scanning, a yield measurement with specified collection and decay times is started. At the end of each measurement cycle the picomotor at the etalon mount

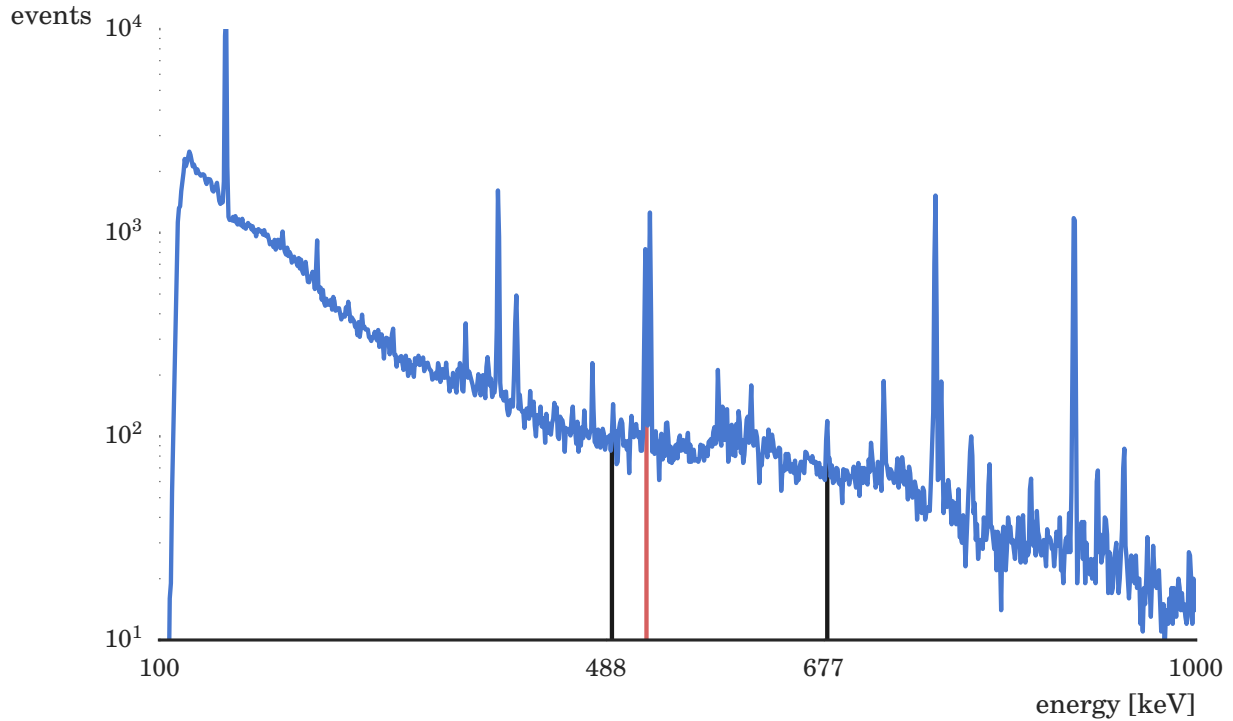


Figure 5.4: γ spectrum recorded with the HPGe detector at the ISAC Yield Station during a ^{118}Ag measurement. The γ energies marked with a black line can be found in both spectra of $^{118g,m}\text{Ag}$. The red line indicates the 511 keV-annihilation peak. The majority of the remaining γ lines visible belong to the isobaric background of ^{118}In .

moves by a specified step size. Unfortunately, the frequency increments from measurement cycle to measurement cycle are not always identical.

In this section, the isotopes discussed were analyzed by evaluating their γ spectra. Both the γ radiation and α particles are recorded with respect to detection time and decay energy. Such a γ spectrum for ^{118}Ag is shown in Figure 5.4. In order to determine the yield for a γ or α emitting isotope, the events detected during the decay time in an isotope-specific energy interval are integrated. Since every isotope decays only once with the same energy, the detected events correspond to the difference of nuclei, ΔN between the start of the decay time interval and the end.

Using eq. (5.2), the isotope difference can be expressed as

$$\Delta N = N_0 - N(t_{\text{decay}}) \quad (5.7)$$

$$= N_0 - N_0 \cdot e^{-\lambda t_{\text{decay}}}, \quad (5.8)$$

where here as well $N_0 = N(t_{\text{collect}})$. By rearranging eq. (5.8), and substituting in eq. (5.3), the activation rate can be written as

$$P = \frac{\lambda}{1 - e^{-\lambda t_{\text{collect}}}} \cdot \frac{\Delta N}{1 - e^{-\lambda t_{\text{decay}}}}. \quad (5.9)$$

In order to determine the yield from this result, the branching ratio b also has to be taken into account, where the branching ratio describes the probability of the nucleus to decay along a certain path. Therefore, the yield from individual, characteristic γ lines can now be calculated by

$$\text{yield} = \frac{P}{\epsilon n b}. \quad (5.10)$$

For a frequency scan the counts in the corresponding γ lines are monitored within a certain counting time represent the isotope yield, since all other parameters are constant. Since the laser frequency changes at the end of every measurement cycle, the Yield Station settings are adjusted that events are also counted during t_{collect} and not only during t_{decay} as is the case for yield measurements. As a consequence, statistics can be improved by extending the implantation time. For yield measurements, especially of short lived isotopes, a longer implantation would not help to increase statistics in the decay time since the activity reaches saturation; see Figure 5.2 where this is the case for the 18 s isomer saturated after approximately 90 s implantation time.

Since the databases [72, 73] for the measured isotopes might be incomplete (e.g., no γ lines are listed for ^{119m}Ag) all γ lines in the spectrum were initially marked, and the counts in these energy intervals were saved according to their cycle number, corresponding to a different laser wavelength

additionally recorded. By plotting the counts in each γ line with respect to the laser wavelength, it is possible to see which line belongs to the ground-, an isomer-, or a background isotope. Those plots for the γ lines that seem to belong to a silver isotope are shown in Figures A.1 to A.4. Afterwards, the counts in the γ lines that could be clearly assigned to either the ground or isomeric state of the corresponding silver isotope were added together. The summarized data was evaluated in a similar way to that described in the section above. The only difference being, that the data was binned to compensate for the nonlinear scanning behavior of the laser. For the spectrum in Figure 5.5 labeled as ^{118m}Ag no γ line was found that did not also appear in the ground state; therefore, the single peak spectrum of the ground state was first fitted. Following this, the spectrum for the isomer was fitted with three Gaussians where the center peak was fixed to the previously evaluated centroid.

5.4 Isotope shift in silver

The IS as defined in section 3.4.2 is the transition frequency difference between two isotopes of the same element. In both Figures 5.3 and 5.5 the frequency shift is given by the difference between the absolute wavelength measured and that listed in the database [8, 9] for ^{109}Ag , as shown in Figure 5.1. In order to find the center frequency for the scans with two distinguishable peaks the CoG had to be determined. For that purpose, the relative intensities of the underlying, unresolved HFS components were calculated by using eq. (3.31). The CoG can now be defined as the weighted mean between the center frequencies of the two Gaussians, where the relative intensities $S(F_g \rightarrow F_e)$ are used as weights

$$\text{CoG} = \nu_{\text{high}} \cdot \sum_i S(F_{\text{glow}} \rightarrow F_{e_i}) + \nu_{\text{low}} \cdot \sum_i S(F_{\text{ghigh}} \rightarrow F_{e_i}). \quad (5.11)$$

Since the HFS splitting of the excited state is much smaller than that of the ground state $A(^2P_{3/2}) \ll A(^2S_{1/2})$, the energy difference from the lower F in the ground state to one of the levels in the excited state is larger than that from the higher F , as it is schematically shown in Figure 5.6. Therefore the

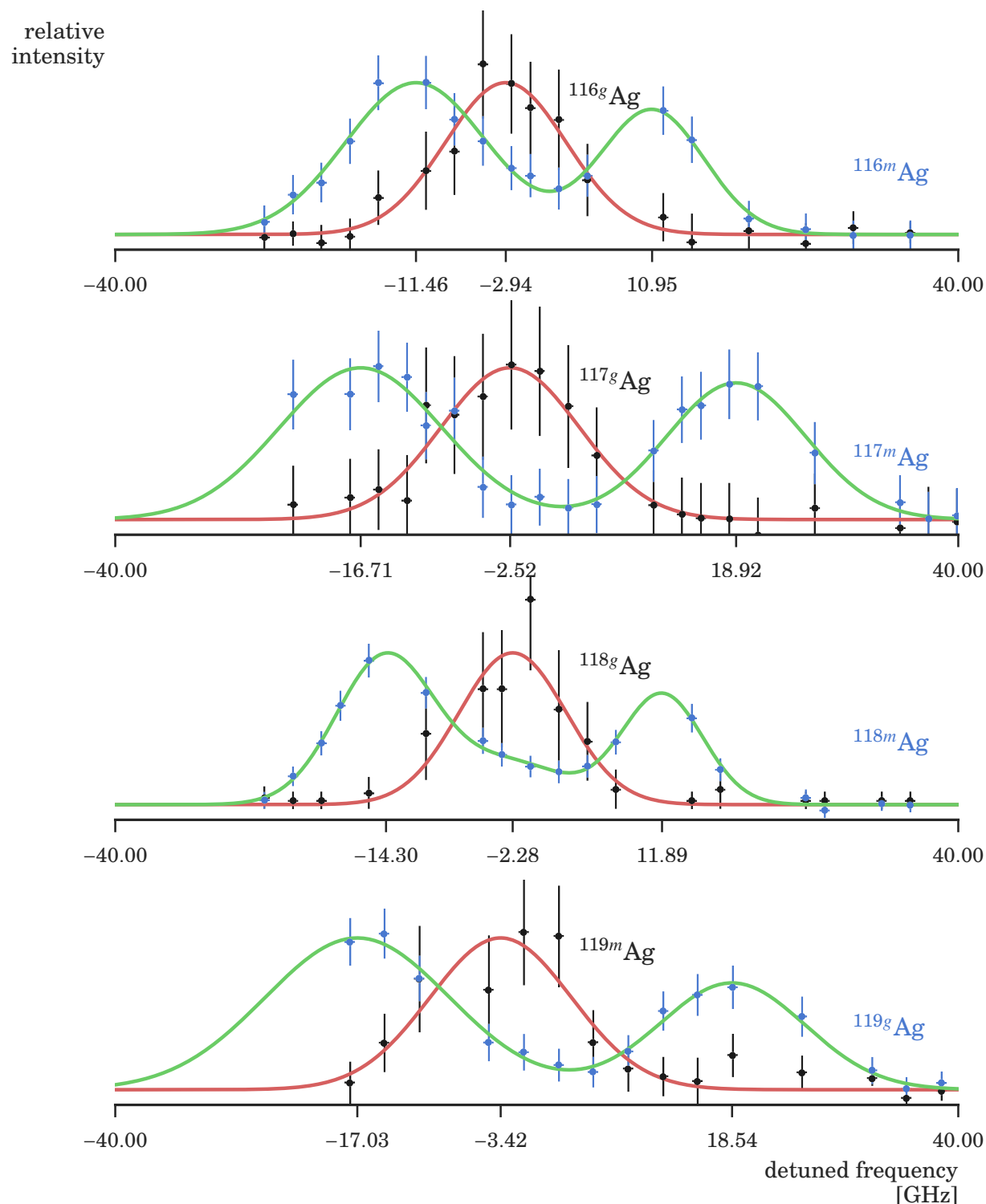


Figure 5.5: Frequency scan of the first excitation step in $^{116-119}\text{Ag}$. The scan frequency is relative to $30\,472.67\text{ cm}^{-1}$. The red curve fits the spectrum of the nuclear ground state, and the green curve that of the isomer. However, the spin assignment for ^{119}Ag is not clear. Furthermore no γ lines were listed for the ^{119m}Ag nucleus. By monitoring the events in each line as a function of the laser frequency, it is possible to assign each γ line to the nucleus of origin.

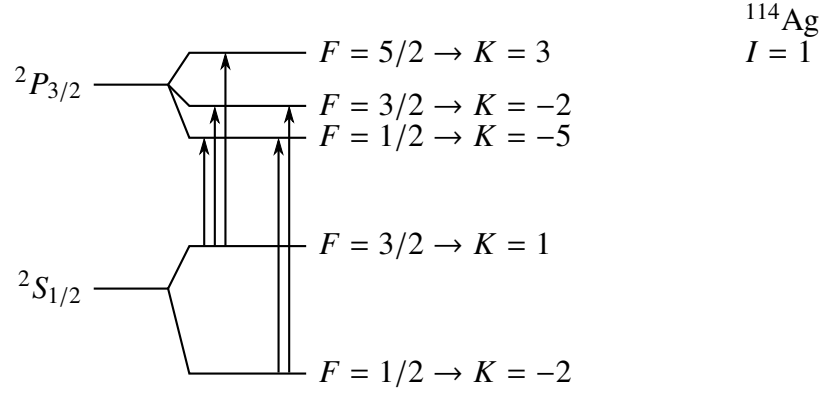


Figure 5.6: Level scheme for ^{114}Ag which has an $I = 1$ nucleus. The atomic ground state splits into two HFS components, the excited state in three. Even though five transitions are possible, with the available spectroscopic resolution only the splitting of the ground state can be resolved. Indicated are total angular momenta F and the corresponding K for each level.

transitions from the lower HFS component can be assigned to the higher frequency peak and vice versa. The results are shown in Tables 5.2 and 5.3.

It can be seen from eqs. (3.31) and (5.11), that the CoG is strongly nuclear spin dependent. This nuclear property, however, is not known for all measured isotopes. Depending on the database ^{116}Ag i.e. is listed with either two isomers of $I_g = (0)$, $I_{m1} = (3)$, and $I_{m2} = (6)$ [72] or with one isomer where the spins are assigned to $I_g = (2)$ and $I_{m1} = (5)$ [73]. Even though a single peak component is present in the spectrum, the first case is not necessary the most likely one, since for ^{112}Ag ($I=2$) a small magnetic moment of $\mu_I = 0.0547(5)\mu_N$ is listed [76], which would also result in a single peak spectrum. Due to the low statistics, it is furthermore not possible to identify if in the double peak spectrum two separate isomer spectra are present. For this isotope the IS was calculated for all possible nuclear spins. Furthermore it should be noted that for both ^{114}Ag and ^{118}Ag the ground state is listed with $I = 1$, where in the case of ^{114}Ag a splitting of $\Delta\nu = 27.7(6)$ GHz was measured, but only one peak was observed in ^{118}Ag . In ^{119}Ag it is not clear which spin belongs to the ground and which to the isomer.

In order to be able to compare the measured data with the literature values of the neutron-deficient isotopes, the change of the mean squared charge radius $\delta\langle r^2 \rangle$ was determined by using eq. (3.52).

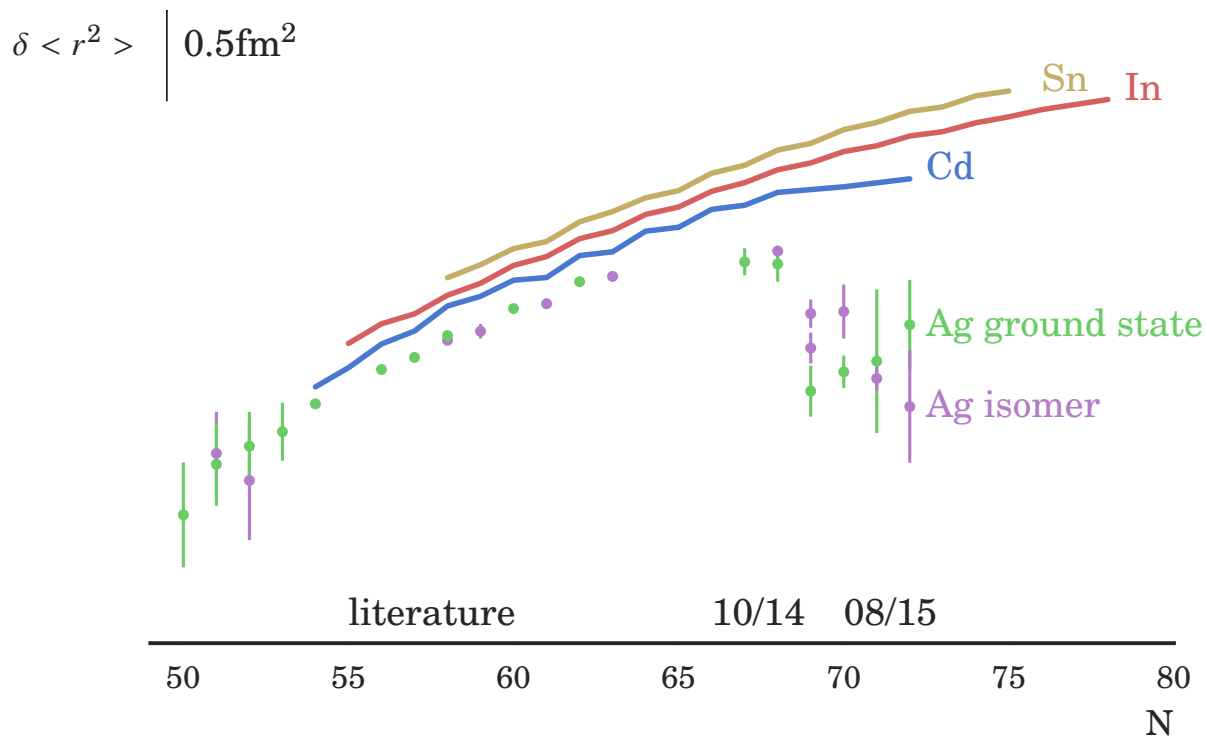


Figure 5.7: Change of the mean squared charge radii of silver isotopes, in comparison to the neighboring isotope chains of cadmium, indium and tin. $^{97-110}\text{Ag}$ are taken from [68, 53], $^{114,115}\text{Ag}$ was measured in October 2014 and $^{116-119}\text{Ag}$ in August 2015. Since no common isotope has been measured between those data sets, the offset seen in the silver isotope chain is most likely due to a systematic error in the measurement. The true effect needs to be determined. The data point of the ground and isomeric state in ^{119}Ag might be reversed as a clear assignment for the nuclear spin of this isotope is not available.

The theoretical electronic factors were collected in [68] and references therein as

$$\lambda^{A,A'} = 0.976 \cdot \delta \langle r^2 \rangle$$

$$F = -4.3(3) \text{ GHz}$$

$$\text{SMS} = \text{NMS} \cdot 0.3(9).$$

The result is shown in Figure 5.7.

The data was collected in two independent experiments approximately one year apart. Although the wavelength meter is routinely calibrated with a frequency stabilized He:Ne laser, it was shown during

the HFS measurements in actinium that the absolute frequency deviated by 800 MHz between the two experiments. The relative frequencies stayed unchanged. In normal laser ion source operation this shift is not critical, since the resonance linewidth is due to broadening mechanisms and the laser linewidth commonly in the order of 10 GHz, and was therefore not noticed. For the measured isotopes no literature values are available. The experiments did not measure identical isotopes. The drastic offset between the data taken in October 2014 and August 2015 might be explained by the uncertainty of the absolute frequency readout of the wavelength meter. As a result no quantitative statement can be taken about the isotope shift and respectively the change of the mean squared charge radius.

5.5 Nuclear dipole moments

The main reason for the large HFS splitting in certain silver isotopes is the distinct and substantial change of the magnetic dipole moment μ . Although with the available data sets the IS is unquantifiable, it is possible to determine the nuclear dipole moment for scans with a double peak spectrum. Since

$$A_e \approx A_g \cdot 0.0186(4), \quad (5.12)$$

only the splitting of the atomic ground state is resolvable. By neglecting the nuclear quadrupole moment, as it cannot be resolved with the available data, the splitting between the two frequency peaks $\Delta\nu$ is directly proportional to the HFS constant A_g of the atomic ground state. Derived from eq. (3.33), the A-factor can be calculated by

$$A_g = \frac{2\Delta\nu}{K(F_{g_{\text{high}}}) - K(F_{g_{\text{low}}})}, \quad (5.13)$$

where K is the coupling constant expressed in eq. (3.34).

Table 5.2: Summary of isotope shifts, magnetic moments, and HFS coefficient for the FES in $^{114-115}\text{Ag}$.

isotope	I	$\Delta\nu$ [GHz]	$\delta\nu_{\text{IS}}^{A,115g}$ [GHz]	$A(^2S_{1/2})$ [GHz]	μ [μ_N]
114	1	27.7(6)	0.1(7)	18.4(4)	2.44(5)
115	1/2		0.0		
115 _m	7/2	38.1(3)	0.3(5)	9.5(1)	4.41(03)

As shown in eq. (3.35), the A -factor is proportional to the magnetic dipole moment. Since the magnetic field $B_J(0)$, and the total atomic angular momentum J are atomic properties and independent of the particular isotope, the magnetic dipole moment can be determine if the remaining quantities are know for a reference isotope

$$\mu = \frac{\mu_{\text{ref}} A I}{A_{\text{ref}} I_{\text{ref}}}. \quad (5.14)$$

In this case ^{109}Ag was selected, which has the reference values of

$$A = -1976.932\,075(17) \text{ MHz} \quad [77]$$

$$\mu = -0.130\,56(2) \mu_N \quad [76]$$

$$I = \frac{1}{2}.$$

For the evaluation of the magnetic moment μ , the nuclear spin I is a critical variable. It was calculated for all possibilities in the ^{116}Ag nucleus. The results from the determination of the IS and the magnetic moment are listed in Tables 5.2 and 5.3.

5.6 Silver spectroscopy summary

The main purpose of this chapter was to introduce the advantages and evaluation process of RADRIS. For this reason the spectra of $^{114-119}\text{Ag}$ have been measured. Silver has the unique

Table 5.3: Summary of isotope shifts, magnetic moments, and HFS coefficient for the FES in silver. For isotopes where the nuclear spin is not known such as $^{116,119}\text{Ag}$, those parameters were calculated for the different possibilities.

isotope	I	$\Delta\nu$ [GHz]	$\delta\nu_{\text{IS}}^{A,116g}$ [GHz]	$A(^2S_{1/2})$ [GHz]	μ [μ_N]
116	(0)		0		
	(2)		0		
116 _m	(3)	22.4(5)	1.0(8)	6.4(2)	2.54(6)
	(5)	22.4(5)	1.6(8)	4.1(1)	2.69(7)
	(6)	22.4(5)	1.8(8)	3.5(1)	2.73(7)
117	1/2		0.4(10)		
117 _m	7/2	35.6(11)	1.8(11)	8.9(3)	4.12(13)
118	1		0.6(21)		
118 _m	4	26.2(4)	0.2(7)	5.8(1)	3.07(4)
119	(7/2)	35.6(18)	1.4(15)	8.9(4)	4.11(20)
	(1/2)		-0.5(17)		

feature that almost every isotope also has a long-lived isomeric state with HFS differences large enough that the ground and isomeric state can be ionized separately from each other. This separation was possible for all isotope except for ^{114m}Ag whose half-life is too short to be extracted from the target.

$^{114,115}\text{Ag}$ was evaluated by monitoring the β decay of those isotopes. Due to the different half-lives of silver compared to the isobaric background of indium, it was possible to determine the center frequencies for each isotope, even though no change of the RIB was detected on the CEM, when lasers were added.

The spectrum of $^{116-119}\text{Ag}$ were analyzed by evaluating the isotope-specific γ energies of each isotope. Although no γ lines were listed for ^{119m}Ag , it was possible to assign seven γ lines to a single peak, and 16 γ lines to the measured double peak spectrum. For this isotope it is unclear whether the ground or the isomeric state have a nuclear spin $I = 1/2$, therefore the γ lines can only be assigned to a nuclear spin, but not to the particular long-lived state in the nucleus. All γ lines that exist in ^{118m}Ag can also be found in its ground state, therefore the spectrum shown for this nucleus in Figure 5.5 represents a spectrum measured on a CEM. Although it can be seen that

the components of this spectrum are not two pure Gaussians, it is not possible to determine the center frequency for the ground state. However, since the ground state has two γ lines that cannot be found in the isomeric state, it is possible to separate the two spectra from each other.

Since the nuclear spin I is not clearly assigned for all isotopes, and no reference isotope has been measured, it was not possible to extract reliably the IS, HFS constant A , and the nuclear dipole moment μ , with the available data.

6 | Actinium

The main interest in actinium at TRIUMF lies in the production of medical isotopes for studies of α emitters in cancer treatment. α particles have a path length of only $50\text{ }\mu\text{m}$ to $100\text{ }\mu\text{m}$ in human tissue, but a mean energy deposition of about $100\text{ keV}/\mu\text{m}$ [78]. These properties allow the treatment of small tumor clusters and micrometastatic diseases while keeping the surrounding cells unaffected providing the α -emitter can be attached to the tumor cells. The most common method to date for treating these diseases is chemotherapy, which attacks all fast-growing cells. Since not all fast-growing cells are cancerous tissue, also healthy tissue will be destroyed by this treatment leading to the familiar side effects. Conversely, targeted alpha therapy (TAT) uses drugs that detect changes in the DNA of the affected cells associated with rapid cell division and target these.

One of the most promising isotopes for TAT is ^{225}Ac due to its long half-life of 10 d and fast decay chain comprising five more α emitters and only three β^- emitters. The daughter nucleus with the longest half-life of 46 min, ^{213}Bi , is a further common isotope for TAT which, due to its short half-life, is difficult to get to the cancerous cell before the ^{213}Bi decays. Therefore ^{225}Ac can additionally also be used as an in vivo generator of ^{213}Bi [79].

Actinium can only be found in nature as traces of uranium and thorium decay products, and its production is expensive, thus knowledge of its chemical behavior is limited. Single photon emission computed tomography (SPECT) is a possible method for studying the medication distribution at different times post-treatment [80], requiring gamma-emitting isotopes. Suitable actinium isotopes considering half-life and decay mode would be ^{224}Ac (2.8 h), ^{226}Ac (29.4 h), and ^{228}Ac (6.2 h).

Alpha-emitting isotopes for research purposes are produced in the ISAC target and collected at an implantation station located directly downstream of the mass separator. The most efficient ion source for actinium is the laser ion source (LIS). As actinium has a large IS and HFS splitting a beam time was scheduled to study these properties for the neutron-rich actinium isotopes with the aim to improve laser ionization and determine the required transition frequencies.

6.1 Actinium production and ionization

Six hours before the start of the scheduled beamtime to measure the HFS and IS of heavy actinium isotopes, the main cyclotron went down for unscheduled repairs. At that point in time, the uranium carbide target had already been bombarded by $1755 \mu\text{A h}$ of protons resulting in the build up of a significant amount of long-lived isotopes inside the target. Instead of canceling the beamtime, it was decided to concentrate on actinium isotopes that, due to their long half-lives and release times, could still be extracted from the target. Retrospectively, it turned out that this was a good decision. On-line, the isobaric background of francium and radium isotopes was later found to be of the same order of magnitude or greater, despite actinium's expected higher production rate in this mass region as shown in Figure 6.1. The main reason for the low efficiency is the slow release time of actinium from the UC_x target material on the order of a day (discussed in section 2.1). For off-line measurements, this long release time becomes an advantage since it increases the time over which actinium is available. In previous experiments, spectroscopy on ^{225}Ac and ^{227}Ac was feasible up to four weeks after the protons were turned off [19]. A list of the isotopes studied and included in this chapter is given in Table 6.1.

In order to enhance release of the isotopes out of the target, it was resistively heated with 450 A corresponding to a temperature of about 2000 K. A standard surface ion source module was used where inside the ionizer tube a thin rhenium foil was placed to enhance, due to its high work function of $W_{Re} = 4.8 \text{ eV}$, the surface ionization for elements with low IPs. According to eq. (2.5)

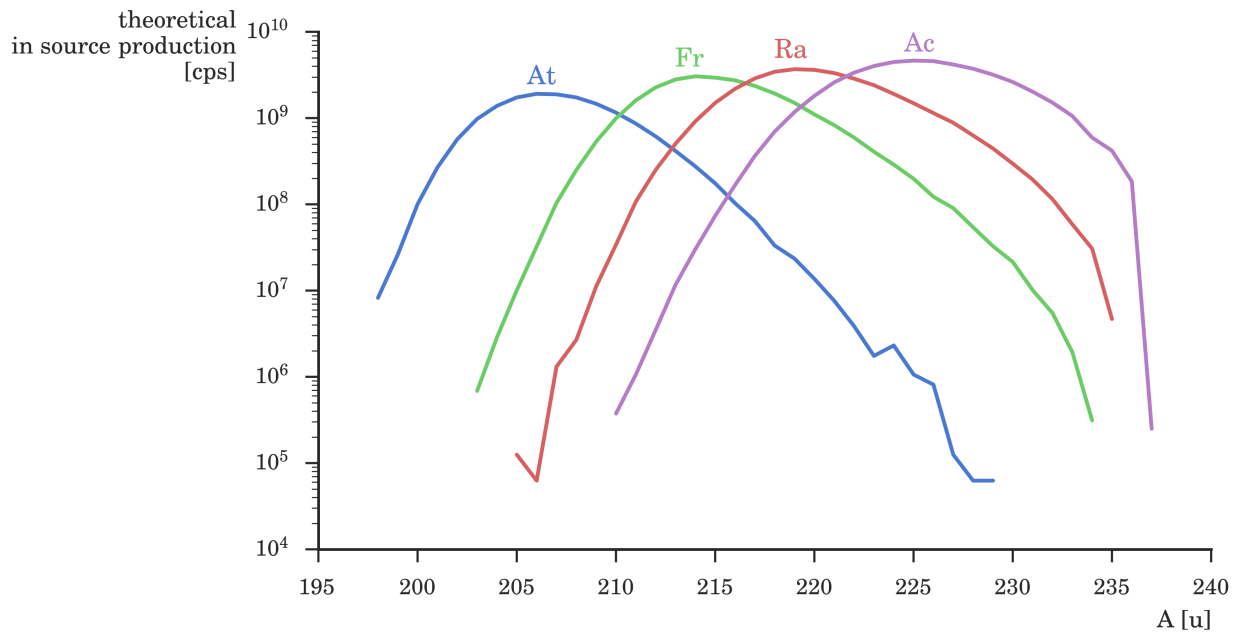


Figure 6.1: Theoretical in-target production for astatine, francium, radium and actinium from a uranium target that is irradiated with $10\mu\text{A}$ of a 500MeV proton beam. The calculations were performed using the GEANT4 software package [81]. Actinium and astatine are the elements of interest. Francium and radium are possible contaminants due to their good surface ionization efficiency of 97 % and 11 %. The mass region of interest is for both elements at $A = 220\text{u}$ and heavier.

Table 6.1: Summary of nuclear decay properties for the measured actinium isotopes. From the half-life of the isobars with sufficient surface ionization, it can be seen that contamination for off-line operation is only expected from the radium isotopes.

nuclide	half-life	spin I	isobaric Fr half-life	isobaric Ra half-life
^{225}Ac	10.0 d	(3/2)	4 min	14.9 d
^{226}Ac	29.37 h	(1)	49 s	1600 y
^{227}Ac	21.77 y	3/2	2.47 min	42.2 min
^{228}Ac	6.15 h	3	38 s	5.75 y
^{229}Ac	62.7 min	(3/2)	50.2 s	4 min

the ionization efficiency of actinium with an $\text{IP} = 5.38 \text{ eV}$ from a rhenium surface at 2000 K is about 0.9 %. An ion current enhancement by a factor of 5.2(5) has been measured on-line by laser ionization. With the protons turned off, a further measurement was performed and the enhancement increased to a factor of 16.0(17), a further advantage for performing actinium spectroscopy on a previously irradiated target. A possible explanation for the difference between these enhancement factors could be, that in on-line operation the power deposit of the proton beam heats in addition to the target container also the ionizer tube and therefore increases the surface ionization efficiency.

Most information available on the atomic energy levels of actinium is known from the emission spectroscopy of arc and hollow cathode lamps using the $t_{1/2} = 21.8$ years long-lived isotope ^{227}Ac [82, 83]. Based on these transitions, a search for suitable AIs was carried out at the Johannes Gutenberg-University (Mainz, Germany) [84, 85] and TRIUMF [19] to find the most efficient excitation scheme for resonant laser ionization and HFS studies. The HFS and IS of actinium was investigated here on two different transitions, 438 nm and 466 nm, using the excitation schemes shown in Figures 6.2 and 6.3. In the atomic spectra database of the National Institution of Standards and Technology (NIST) [8], the lines are listed as transitions from the atomic ground state $6d7s^2 \ ^2D_{3/2}$ to $6d7s(^3D)7p \ ^4P_{3/2}$ and $6d7s(^3D)7p \ ^4P_{1/2}$ state, respectively. From the overlap of common AIs in the above mentioned experiments, it has been shown that the assignment of the total angular momentum is erroneous in both cases. Even though the 438 nm is a transition from the atomic ground state, the total angular momentum of the excited state was assigned to be

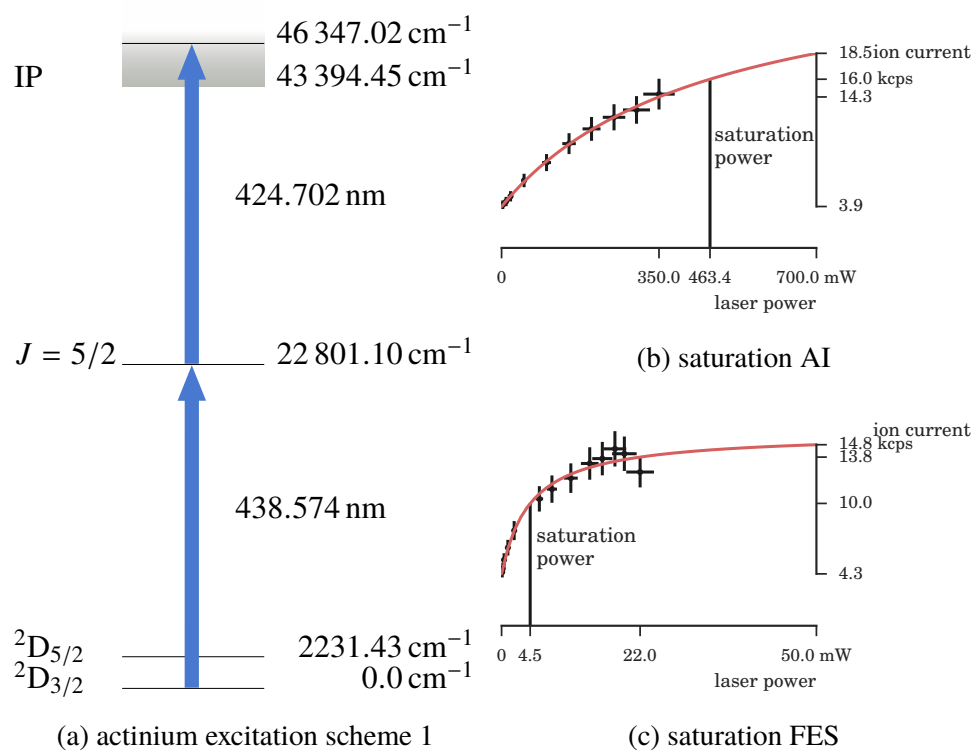


Figure 6.2: Actinium excitation scheme 1 with laser power saturation curves for the two resonant excitation steps.

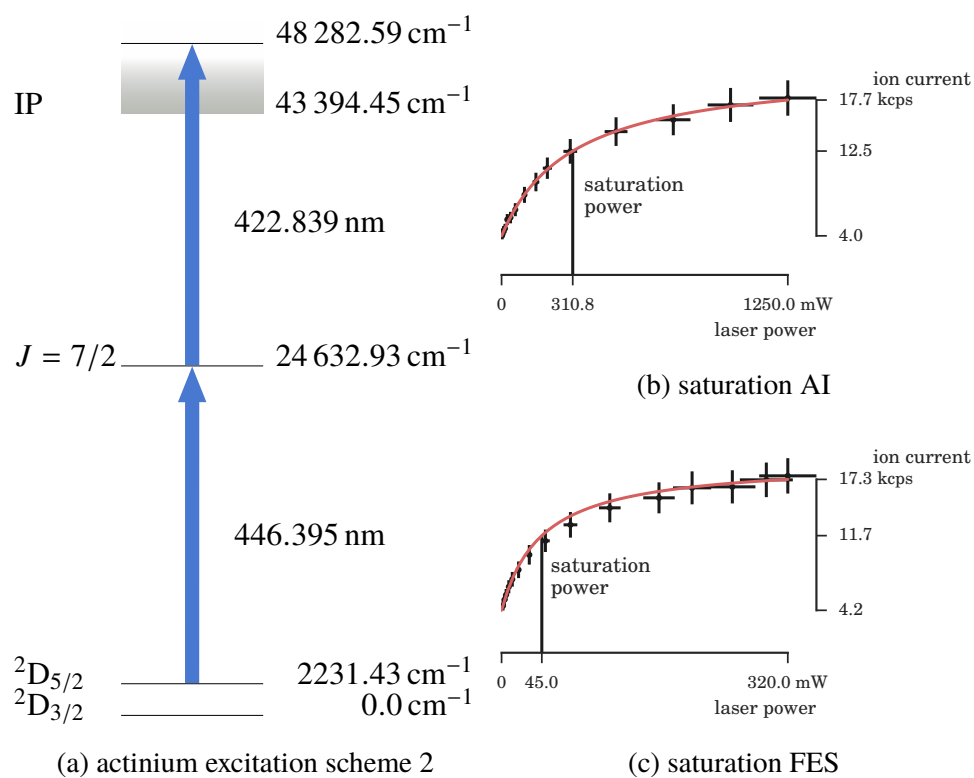


Figure 6.3: Actinium excitation scheme 2 with laser power saturation curves for the two resonant excitation steps.

$J = 5/2$ [86]. The second excitation scheme starts from the $6d7s^2\ ^2D_{5/2}$ low-lying excited state of 2231.43 cm^{-1} , which is thermally populated by approximately 30 % at 2000 K as demonstrated in section 3.1.2. The excited state of this transition has a total angular momentum of $J = 7/2$ [87].

Except for the AI in *excitation scheme 1* that is presented in Figure 6.2, all transitions were saturated with the laser power available during the measurements. The saturation curves were recorded for ^{225}Ac on CEM, an electron multiplier located just downstream of the mass separator, with a 10 times attenuation of the radioactive ion beam. At first glance, *excitation scheme 2* seemed to be more promising for efficient laser ionization since higher ion currents were achieved. However, if for both schemes the same laser power would have been available, the ion current for *excitation scheme 1* is expected to be higher than for *excitation scheme 2*. In addition, based on the large HFS splitting in the first excited state, a broad autoionizing step is advantageous in being able to ionize all HFS components simultaneously. As a result of the substructure in the AI in *excitation scheme 1*, this transition is one order of magnitude broader than that of *excitation scheme 2* as shown in Figure 6.4.

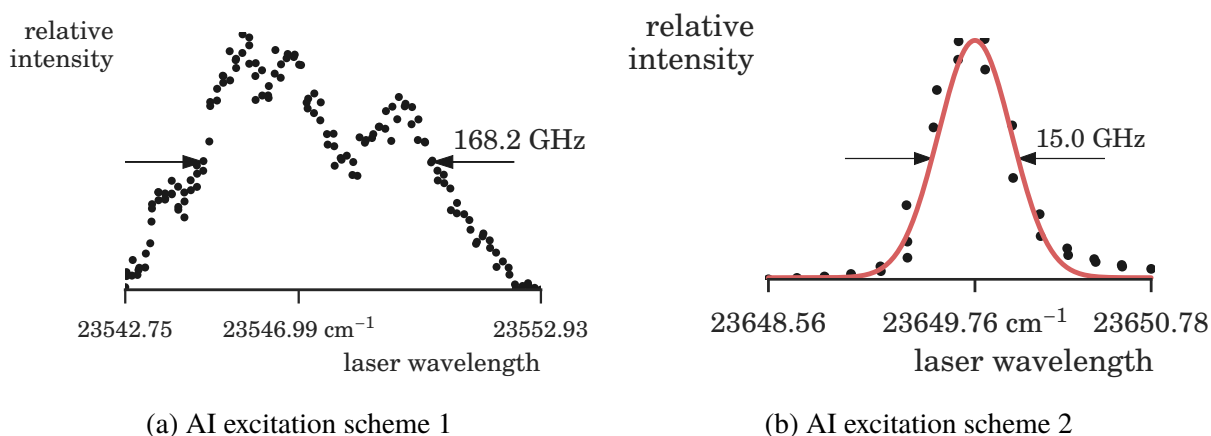


Figure 6.4: Autoionization states in Ac for the two excitation schemes used for laser ionization.

Both excitation schemes use two resonant transitions in the blue spectral region. These wavelengths were generated by internal doubling of the laser light as described in section 4.2. A dichroic mirror was placed inside the resonator in order to double the laser output power but this also obstructed an otherwise continuous scan. As shown in the example of Figure 6.4b, the measured data points are

grouped with a separation of 4.7(3) GHz. For a plano etalon this corresponds to a mirror spacing of 32(2) mm, which agrees with the spacing between the output coupler and the dichroic mirror in the resonator.

6.2 Hyperfine structure and isotope shift

For most efficient laser ionization, each excitation step needs to be fully saturated. On the other hand, fully saturated transitions are broadened due to power broadening, as described in section 3.3.2. In order to measure the HFS of the transition, a compromise has to be found between ionizing sufficient atoms for sufficient statistics, and reducing the laser power to resolve the HFS. The FES scans of *scheme 1* for ^{225}Ac for different laser powers in the first and ionizing step are shown in Figure 6.5. Although the ion current is reduced by an approximate factor of 50, the four HFS components of the excited state, which has the largest splitting as shown in Table 6.2, are only resolved when in addition to the FES the ionizing step is also reduced well below the saturation power. For that reason, whenever the ion current was sufficiently high during a scan, the laser power for both transitions was reduced.

The HFS of ^{227}Ac had previously been studied for five different transitions at the Johannes Gutenberg-University (Mainz, Germany) using a narrowband injection-locked Ti:Sa laser system developed at the University of Jyväskylä [88]. The results from these measurements for the energy levels used in this work are listed in Table 6.2. In order to fit the HFS of the measured scans,

Table 6.2: HFS coefficients A , B for transitions in actinium used in this thesis, taken from [88].

energy level [cm^{-1}]	A [MHz]	B [MHz]
0.00	50.5(4)	595.6(21)
2231.43	255.0(13)	706.6(78)
22 801.10	2104.8(5)	110.1(22)
24 632.93	NA	NA

these HFS coefficients were used as starting values. Since the atomic properties remain unchanged

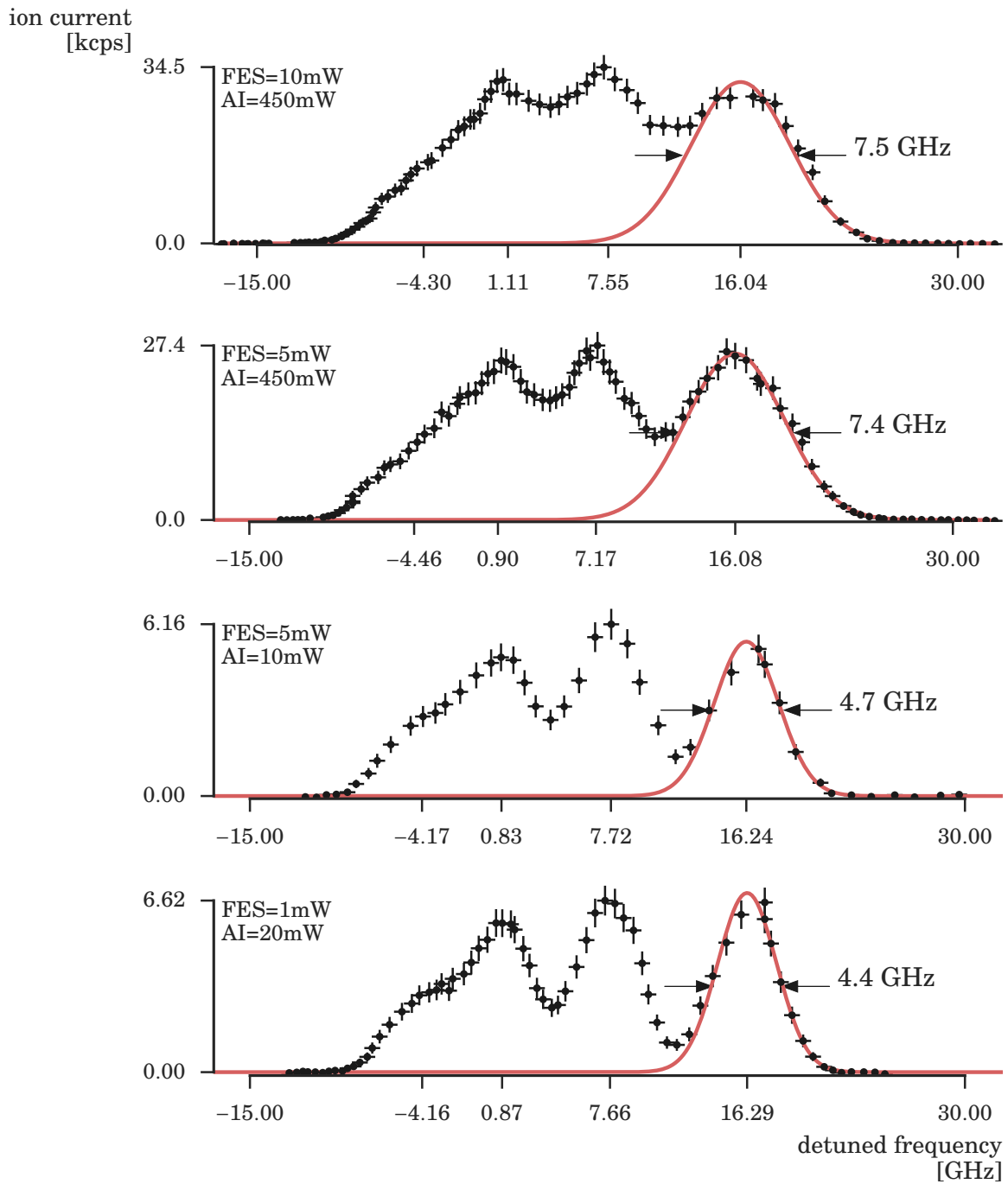


Figure 6.5: First excitation step scans for the excitation scheme shown in Figure 6.2 on ^{225}Ac . The laser power of the first and ionizing step is varied to find the compromise between efficient laser ionization, and resolution. The small HFS component -4 GHz away from the ^{227}Ac center-wavelength at $22\,801.1\text{ cm}^{-1}$ is only resolved for the lower two scans, showing that not only the laser power of the FES but also that of the ionizing step contributes to the power broadening. The peak at 16 GHz contains only one HFS component and is therefore fitted with a single Gaussian to test for power broadening. In addition the centroids of all four distinguishable peaks are indicated.

for the different isotopes, the ratio between the HFS coefficients of the ground and excited states $\frac{A_e}{A_g} = \frac{2104.8(5)}{50.5(4)} = 41.7(3)$ and $\frac{B_e}{B_g} = \frac{110.1(22)}{595.6(21)} = 0.185(4)$ are kept constant during the fit. The peak amplitude was fitted using the theoretical relative intensities (eq. (3.31)) and assigning every distinguishable peak a separate fitting amplitude. Since the laser pulse length is on the order of the lifetime of the excited atomic state, optical pumping within the HFS is neglected. For every isotope between 5 and 15 scans were conducted. Each of these scans was fitted individually and afterwards a weighted average of the fit results was taken. The outcome of these fits is summarized in Table 6.3.

All isotopes except ^{226}Ac were measured on a CEM using the Yield Station program for detuning the laser frequency. For each measurement interval with the same laser frequency, the ion counts on the CEM were summed up. ^{226}Ac was measured directly at the Yield Station because the long-lived isobaric radium background was too high to identify the resonant laser-ionized actinium signal on the CEM. Although ^{226}Ac has a detectable α - as well as a γ -spectrum, the β decay branch is the strongest. Therefore, only the scintillator signals were used to evaluate the scans. Despite the fact that the scintillator cannot resolve any nucleus-specific properties except for half-life, this signal is contamination-free since ^{226}Ra is a pure α emitter.

^{228}Ac has a nuclear spin of $I = 3$ and therefore a spectrum with 12 HFS components. Due to the larger splitting in the excited state compared to the splitting in the ground state, six distinguishable peaks are expected as demonstrated in Figure 6.6. In reality, however, these six peaks were not resolvable. Nevertheless, in order to extract the CoG, it was assumed, since the contribution of the spectroscopic A -factor is much larger than the B -factor, that the latter is negligible in the fit. Furthermore, the Gaussian linewidth was fixed to $\sigma = 1.33(3)$ GHz which corresponds to a FWHM = 3.12(6) GHz. This linewidth was extracted from the fits of the other HFS scans taken during the same run. In comparison, the Doppler width for this transition lies at $\Delta\nu_D = 1.48$ GHz. While the A -factor is largest in the excited state, in the ground state the B -factor is the dominant

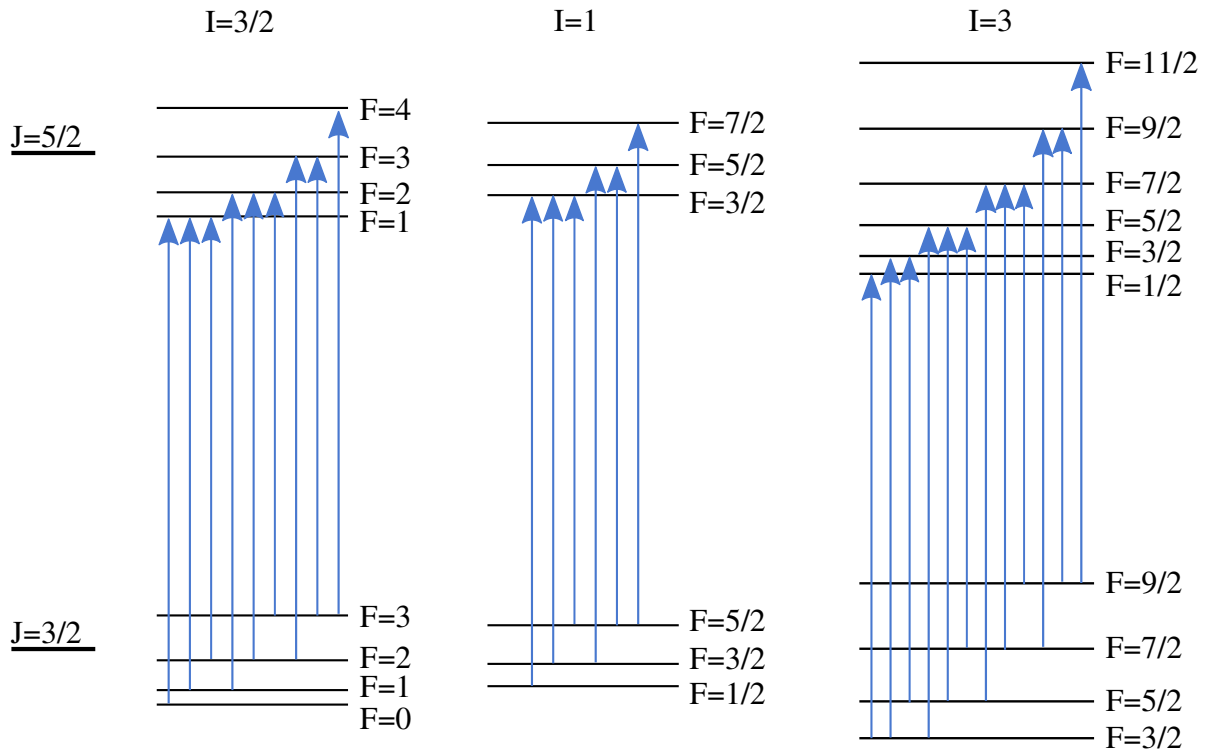


Figure 6.6: Actinium HFS level schemes for the first transition in *excitation scheme 1*. The level splitting is shown for nuclei with $I = 3/2, 1, 3$.

contribution to HFS splitting. Consequently, the real A -factor is expected to be smaller than that extracted from this fit.

Table 6.3: Results of the HFS fits for *excitation scheme 1*.

Isotope	$\delta\nu_{\text{IS}}^{A,227}$ [GHz]	A_g [MHz]	B_g [MHz]	A_e [MHz]	B_e [MHz]
225	7.97(8)	54.3(6)	662.3(1101)	2264.7(269)	122.4(204)
226	3.61(9)	66.6(17)	441.0(722)	2774.5(721)	81.5(134)
227	0.00	50.3(4)	661.8(841)	2095.5(178)	122.3(156)
228	-2.59(15)	<24.6(5)		<1024.3(415)	
229	-6.91(11)	75.5(11)	510.9(274)	3145.4(248)	94.4(51)

Despite *excitation scheme 1* being more efficient for laser ionization, *excitation scheme 2* is expected to have a larger spectroscopic B -factor which helps to extract the electric quadrupole moment and hence the deformation of the nucleus. For this reason, *excitation scheme 2* is of higher interest for nuclear structure studies. Excitation scheme 2 was tested towards the end of the experiment when only two isotopes, ^{227}Ac and ^{225}Ac , were still accessible due to their long half-lives. For this excitation scheme, only the HFS constants A and B for the ground state of the transition were available. In order to determine the HFS constants for the excited state, the scans for ^{227}Ac were fitted by keeping $A_g = 255.0(13)$ MHz and $B_g = 706.6(78)$ MHz fixed and only varying the constants of the excited state. To prove the validity of this method, the same procedure was performed for the *excitation scheme 1* scans of ^{227}Ac and results are shown in Table 6.4. The values extracted from the fits of *excitation scheme 1* are within uncertainties in agreement with the literature value.

Table 6.4: Fitting result of ^{227}Ac to determine the A - and B -factors of the excited states. The fitting results are compared with the literature values [88].

scheme	A_e [MHz]	A_e [MHz] reference	B_e [MHz]	B_e [MHz] reference
1	2095.2(148)	2104.8(5)	80.2(667)	110.1(22)
2	1508.5(5)		1978.3(588)	

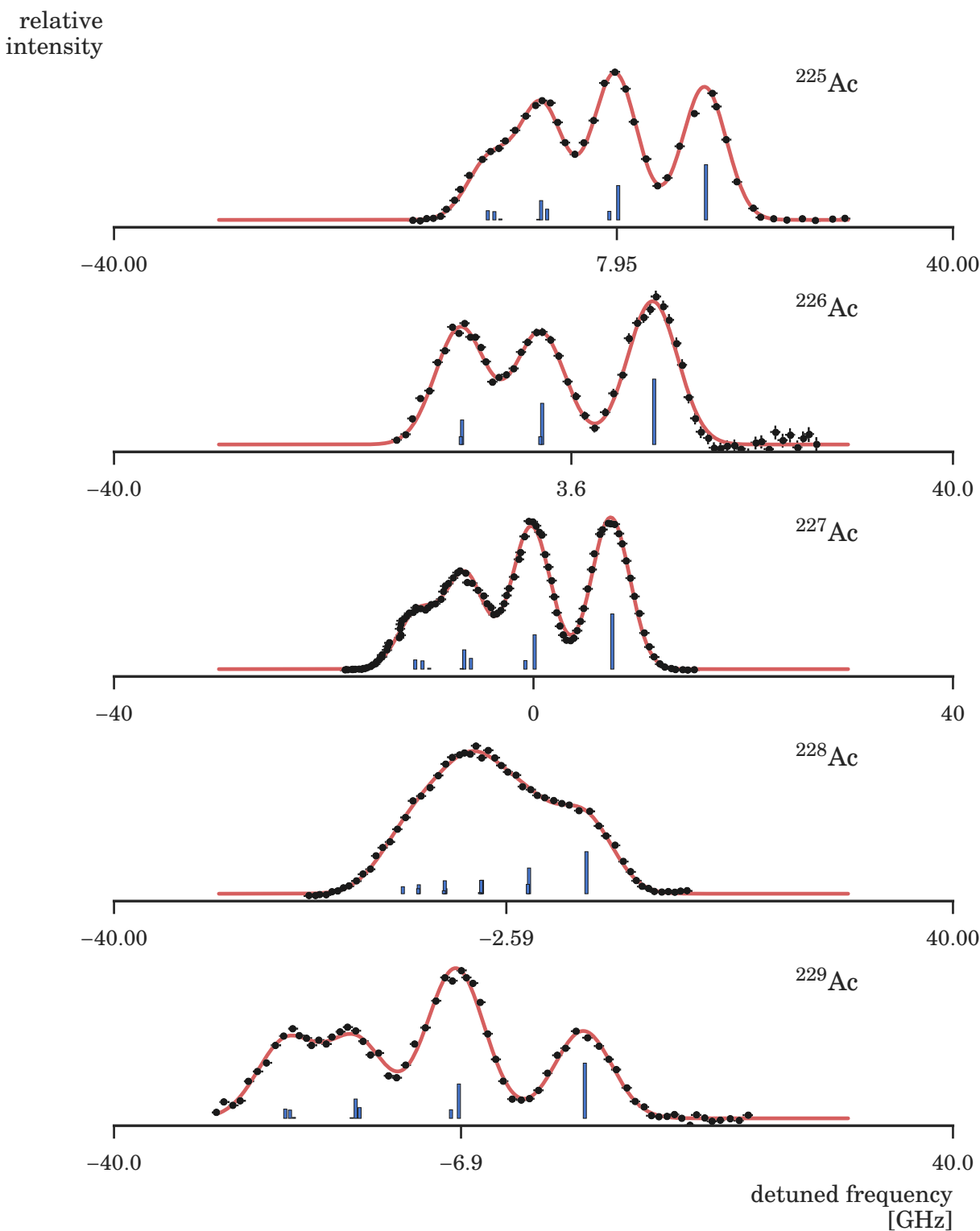


Figure 6.7: Scans of the first transition in *excitation scheme 1* for $^{225-229}\text{Ac}$. The isotope shift relative to ^{227}Ac is marked on the abscissa. The blue bars indicate the underlying HFS components resulting from the fit.

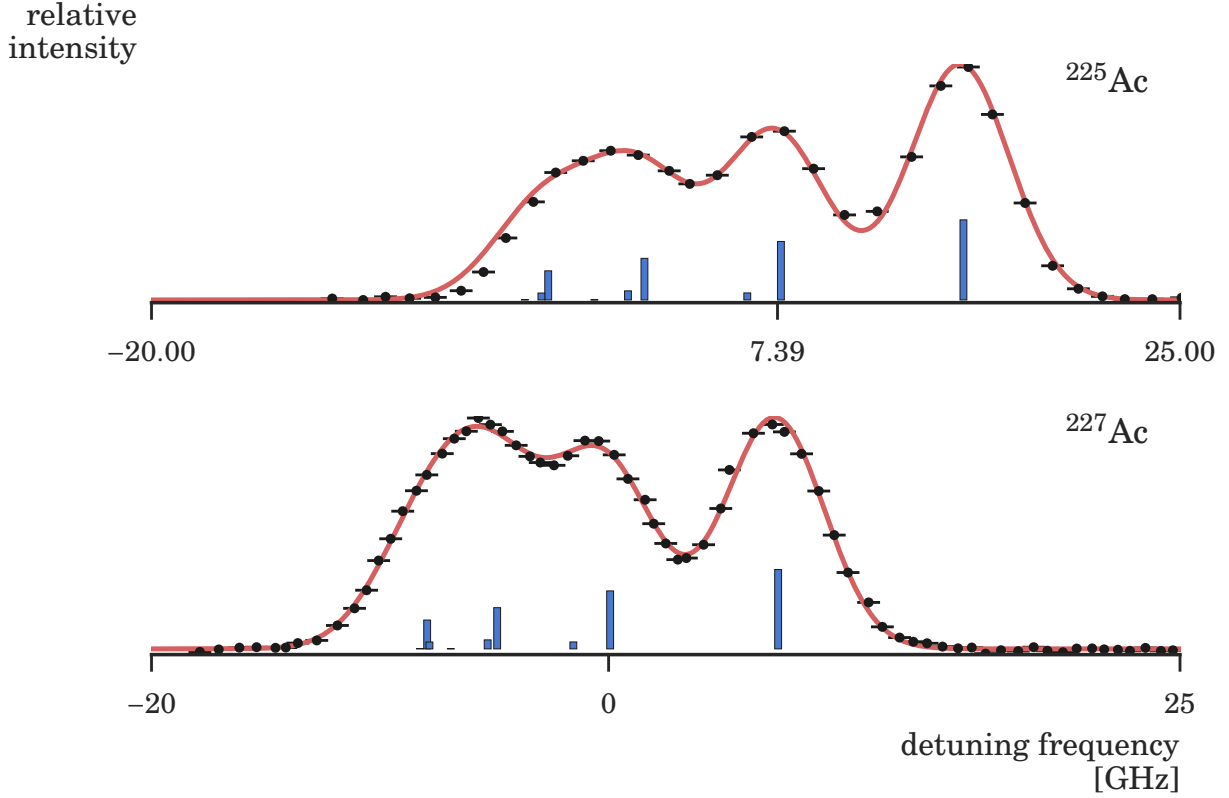


Figure 6.8: Scans of the first transition in *excitation scheme 2* for $^{225,227}\text{Ac}$. The isotope shift relative to ^{227}Ac is marked on the abscissa. The blue bars indicate the underlying HFS components resulting from the fit with their relative intensities.

For the fit of the ^{225}Ac scans of *excitation scheme 2*, again the ratio, between the HFS constants of the ground and excited states $\frac{A_e}{A_g} = \frac{1508.5(5)}{255.0(13)} = 5.92(3)$ and $\frac{B_e}{B_g} = \frac{1978.3(588)}{706.6(78)} = 2.80(83)$ were fixed. The fit results are listed in Table 6.5.

Table 6.5: Results of the HFS fits for *excitation scheme 2*.

Isotope	$\delta\nu_{\text{IS}}^{A,227}$ [GHz]	A_g [MHz]	B_g [MHz]	A_e [MHz]	B_e [MHz]
225	7.39(7)	280.3(42)	530(220)	1659.4(251)	1490(620)
227	0.00	255.0(13)	706.6(78)	1508.5(145)	1980(610)

6.3 Nuclear properties

The nuclear ground state properties extracted from the measured HFSs and ISs is discussed in the following sections.

6.3.1 Nuclear spin of ^{226}Ac

While the nuclear spin $I = 3/2$ of ^{227}Ac is well known as assigned from the HFS of 15 different transitions [89], the spins for $^{225,229}\text{Ac}$ also with $I = 3/2$ and ^{226}Ac with $I = 1$ are still listed in parentheses [72] meaning that the spin is only assigned with weak arguments. Since ^{226}Ac is an odd-odd nucleus, only integer spins are possible. Considering that the HFS spectrum has only three components, the nuclear spin is confirmed to be $I = 1$; as for $I < J_e$, the number of distinguishable peaks is defined by $2I + 1$. For nuclear spin $I > J_e$ six peaks are expected, as is the case for ^{228}Ac but the resolution is not high enough to be actually resolvable as shown in Figure 6.7.

6.3.2 Nuclear moments

The magnetic dipole moment for the ground state of only two actinium isotopes ^{227}Ac and ^{217}Ac , can be found in the literature so far [76]. The HFS components A and B are directly proportional to the magnetic dipole and the electric quadrupole moments respectively. Assuming the magnetic field gradient $B_J(0)$ and electric field $\langle \frac{\partial^2 V}{\partial z^2} \rangle$ at the nucleus are constant across all isotopes for the same atomic states, the nuclear moments can be extracted by using eqs. (3.35) and (3.37) as follows

$$\mu = \frac{\mu_{\text{ref}} A I}{A_{\text{ref}} I_{\text{ref}}} \quad \text{and} \quad Q_s = \frac{B Q_{s\text{ref}}}{B_{\text{ref}}}, \quad (6.1)$$

where the index "ref" refers to the parameters of the reference isotope; in this case, ^{227}Ac . The nuclear moments extracted from the HFS components are listed in Table 6.6. For ^{225}Ac the spectra

Table 6.6: Nuclear moments calculated from the HFS constants extracted from laser spectroscopy using two excitation schemes

Isotope	μ [μ_N]	Q_s [b]	ref.
225 (from scheme2)	1.18(20)	1.3(7)	this work
225 (from scheme1)	1.15(19)	1.93(34)	this work
226	0.94(16)	1.29(22)	this work
227	1.07(18)	1.74(10)	[90]
228	<1.04(18)		this work
229	1.60(27)	1.49(12)	this work

from two excitation schemes were available, the nuclear moments were also extracted for *excitation scheme 2*. Table 6.6 shows that the magnetic dipole moments are in good agreement with each other, while electric quadrupole moments lie within the errors. Due to the large errors for the electric quadrupole moment, the only statement that can be given regarding its deformation is that the nuclei trend towards a prolate shape. Determination of the quadrupole moments requires a high resolution spectroscopy technique such as collinear fast beam laser spectroscopy with narrow bandwidth continuum wave lasers [91]. By comparing the magnetic dipole moments of the three $I = 3/2$ isotopes $^{225,227,229}\text{Ac}$, it appears that the moment for the ^{229}Ac isotope deviates from the other two. On the other hand, the magnetic moments of these nuclei agree very well with those of the francium isotopes with same neutron numbers (Figure 6.9).

6.3.3 Nuclear charge radius

In this heavy mass region, the main contribution to the isotope shift is from the field shift F , which directly corresponds to the change of the mean squared charge radius $\delta \langle r^2 \rangle$. Under the assumption, that the mass shift, and Seltzer moments can be neglected, eq. (3.52) simplifies to

$$\delta \langle r^2 \rangle \approx \frac{\delta v^{A,A'}}{F}. \quad (6.2)$$

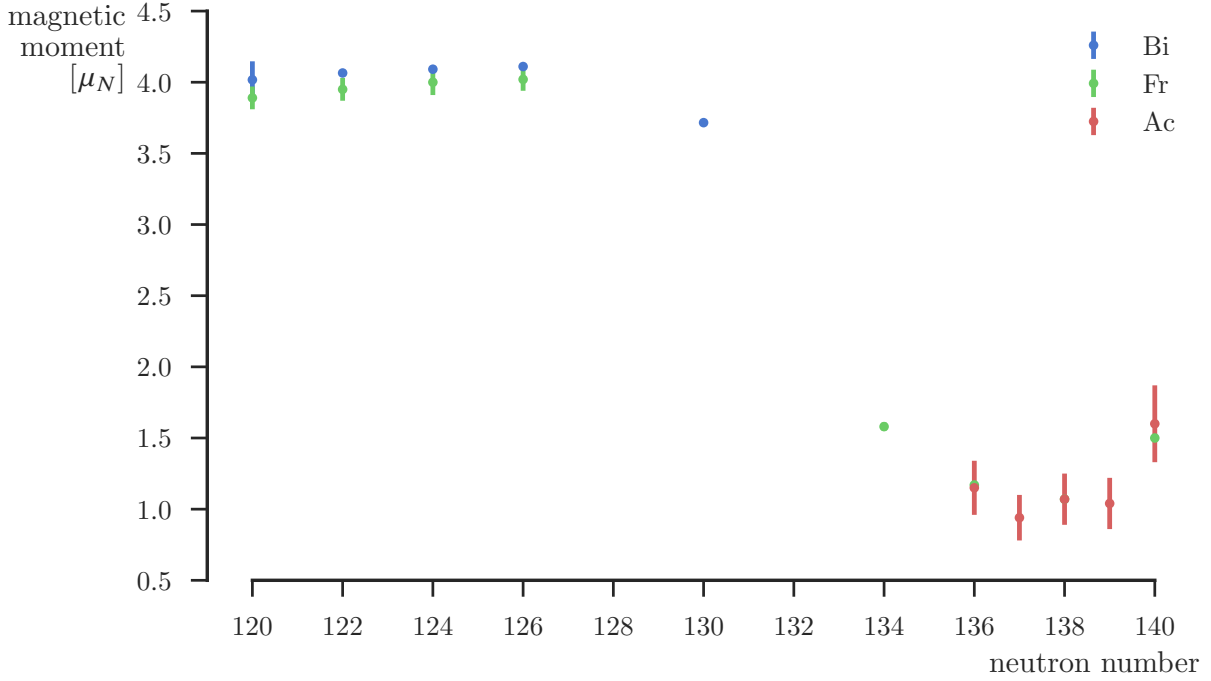


Figure 6.9: Nuclear magnetic moments for odd-even nuclei above lead. The moments for Fr and Bi are from [76]. For completeness, also the dipole moments for odd-odd actinium nuclei are shown.

For the transition used in *excitation scheme 1*, the mass-, and field shift parameters were provided as $M = 500(180)$ GHz u and $F = -39(2)$ GHz fm⁻² by the group of S. Fritzsche (University of Jena, Germany) using ab-initio calculations [90]. The calculated values for $\delta \langle r^2 \rangle$ using eq. (6.2) are provided in Table 6.7. Comparing the calculated $\delta \langle r^2 \rangle$ of actinium with the isotope chains

Table 6.7: Change of the mean squared charge radii of actinium derived from the optical isotope shift measurement of the first excitation step in *excitation scheme 1*.

Isotope	$\delta \langle r^2 \rangle^{A,227}$ [fm ²]
225	0.204(11)
226	0.093(5)
227	0
228	-0.066(5)
229	-0.177(10)

of the neighboring elements, it can be seen in Figure 6.10 that the calculated $\delta \langle r^2 \rangle$ are in good agreement with those of francium and radium.

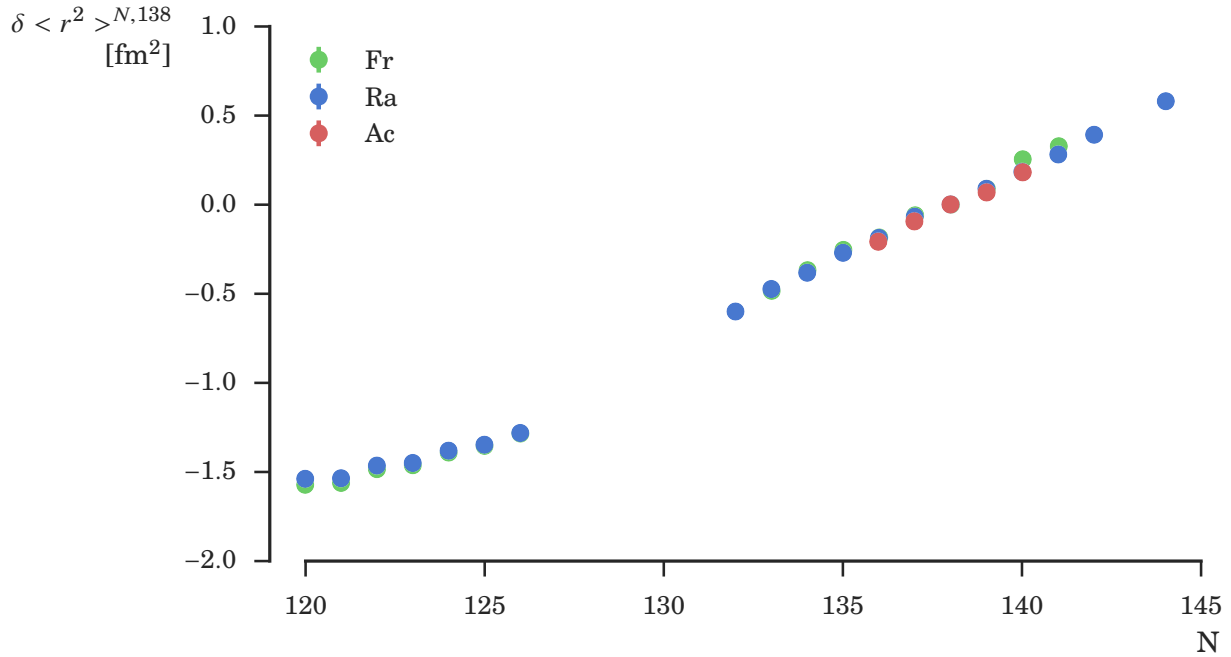


Figure 6.10: Change of the nuclear mean squared charge radii $\delta \langle r^2 \rangle$ of actinium, compared with those of the neighboring isotope chains of francium and radium taken from [11].

6.4 Actinium spectroscopy summary

The HFS and IS for neutron-rich actinium isotopes $^{225-229}\text{Ac}$ have been measured on an activated target without proton irradiation. Due to the long half-lives, the efficiency of measuring the decay products would have been very low. For that reason, the frequency-dependent change of the RIB was for most isotopes directly measured on a CEM downstream the mass separator. Two different excitation schemes have been tested for $^{225,227}\text{Ac}$. The frequency scans for the remaining isotopes were only performed with the most efficient excitation scheme. From the optical spectra, the IS for the FES $\approx -3.73(19) \text{ GHz u}^{-1}$, and the nuclear magnetic dipole moment μ were extracted. For ^{226}Ac the nuclear spin $I = 1$ was confirmed.

7 | Astatine

The main interest in astatine at TRIUMF is the production of neutron-rich isotopes to study the decay into radon. It is expected that the odd-mass $^{221,223}\text{Rn}$ nuclei show an enhanced octupole deformation, which would allow the investigation of a possible permanent electric dipole moment (EDM) [92]. So far a nonzero permanent EDM has not been observed, but the existence of such an EDM would violate both the parity and time-reversal symmetry. The most precise EDM measurement to date has been performed with ^{199}Hg , and its limit confined to $|d(^{199}\text{Hg})| < 7.4 \times 10^{-30} e \text{ cm}$ [93]. Octupole-deformed radon nuclei are predicted to be up to 600 times more sensitive to CP-violating interactions than ^{199}Hg [92]. The majority of data discussed in this work were collected during the RIB delivery for this experiment¹.

7.1 Excitation scheme and ion beam production

Astatine is the least abundant element on earth, mainly because the three isotopes $^{215,218,219}\text{At}$ that are part of natural decay chains have half-lives of only $t_{1/2} = 0.1 \text{ ms}$, 1.5 s and 56 s , respectively [94]. On the other hand, the longest-lived isotope ^{210}At has a half-life of $t_{1/2} = 8.1 \text{ h}$. Therefore, little information about the atomic spectrum was available until recently. The first and only published optical spectroscopy on astatine was performed in 1964 where two transitions from the atomic ground state were discovered [95]. As part of my master thesis [96] I developed a resonant

¹S929: Octupole Deformation and Spin-Exchange Polarization of Odd A Radon Isotopes: toward Radon Electric Dipole Moment Measurements, available at: <https://mis.triumf.ca/science/experiment/view/S929>

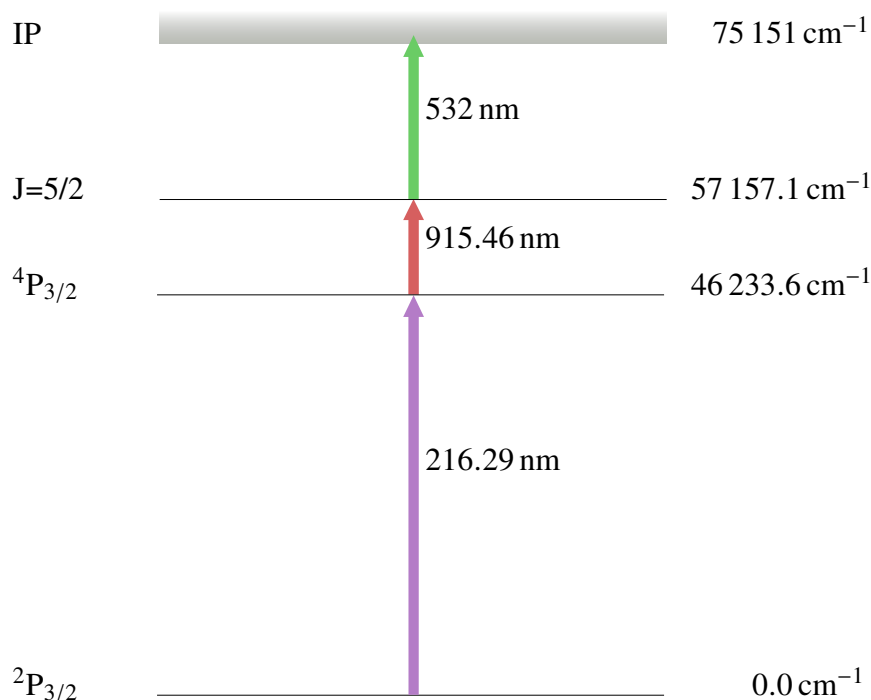


Figure 7.1: Excitation scheme for astatine laser ionization used for resonance ionization in this work.

ionization scheme for astatine, starting with these two transitions. As a result, a total number of 55 new transitions to 41 energy levels were discovered. At CERN the investigation was extended to determine the first ionization potential of astatine via a converging Rydberg series [97]. The most efficient ionization scheme based on this research is shown in Figure 7.1.

Except for the ionizing step, which is realized nonresonantly with the frequency-doubled ionization laser, all excitation steps were saturated with the available laser power. The saturation curves for each step are shown in Figure 7.2. The FES was saturated with $P_{\text{sat}} = 4.0(9)$ mW and the SES with $P_{\text{sat}} = 21(10)$ mW, where for the latter a laser power $P_{\text{laser}} \approx 2$ W was available.

Similar to the other two elements discussed so far, the data for astatine were collected during two independent measurement periods. Most of the FES scans were collected during the beam delivery of ²²¹At to an experiment² aiming to study its γ spectrum. In the requested mass region, the production rates of the possible surface-ionized francium and radium isobaric background are

²S929: Octupole Deformation and Spin-Exchange Polarization of Odd A Radon Isotopes: toward Radon Electric Dipole Moment Measurements, available at: <https://mis.triumf.ca/science/experiment/view/S929>

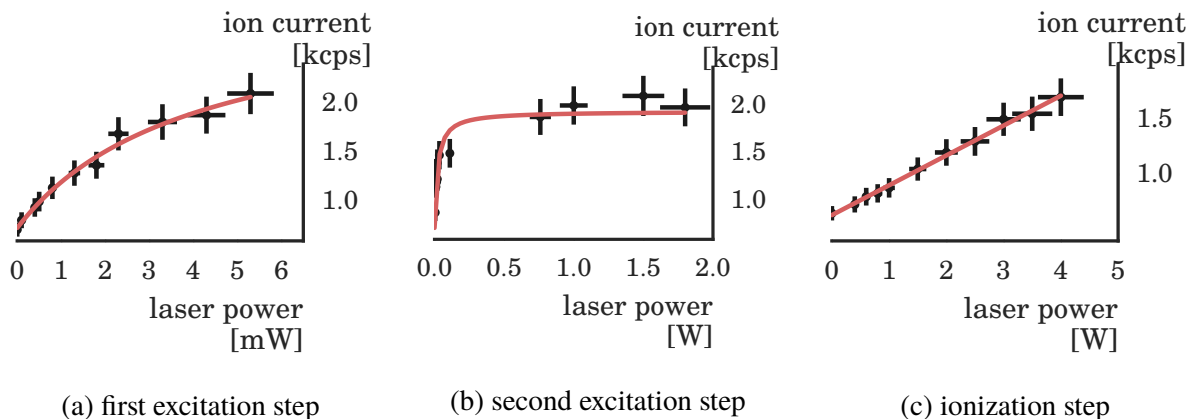


Figure 7.2: Saturation curves for the three excitation steps in the astatine excitation scheme. Both resonant excitation steps are saturated with the available laser power.

expected to be two orders of magnitude higher than for astatine as shown in Figure 6.1. For that reason, the experiment was scheduled to use a UC_x -target in combination with an IG-LIS target module. That way it was possible to reduce the ^{220}Fr background from an average of 4.3×10^6 cps with common surface ion source target modules to less than 100 cps.

The second data set was collected during the test of the first ThO-target irradiated at ISAC. It was planned to study the production of neutron-rich actinium isotopes from this target which were expected to be higher than from a UC_x -target, but no actinium isotopes were released. It was quickly decided to switch to the more volatile astatine to study the performance of this target. Most of the scans of the SES were executed on this target to address inconsistencies in my previously collected data sets.

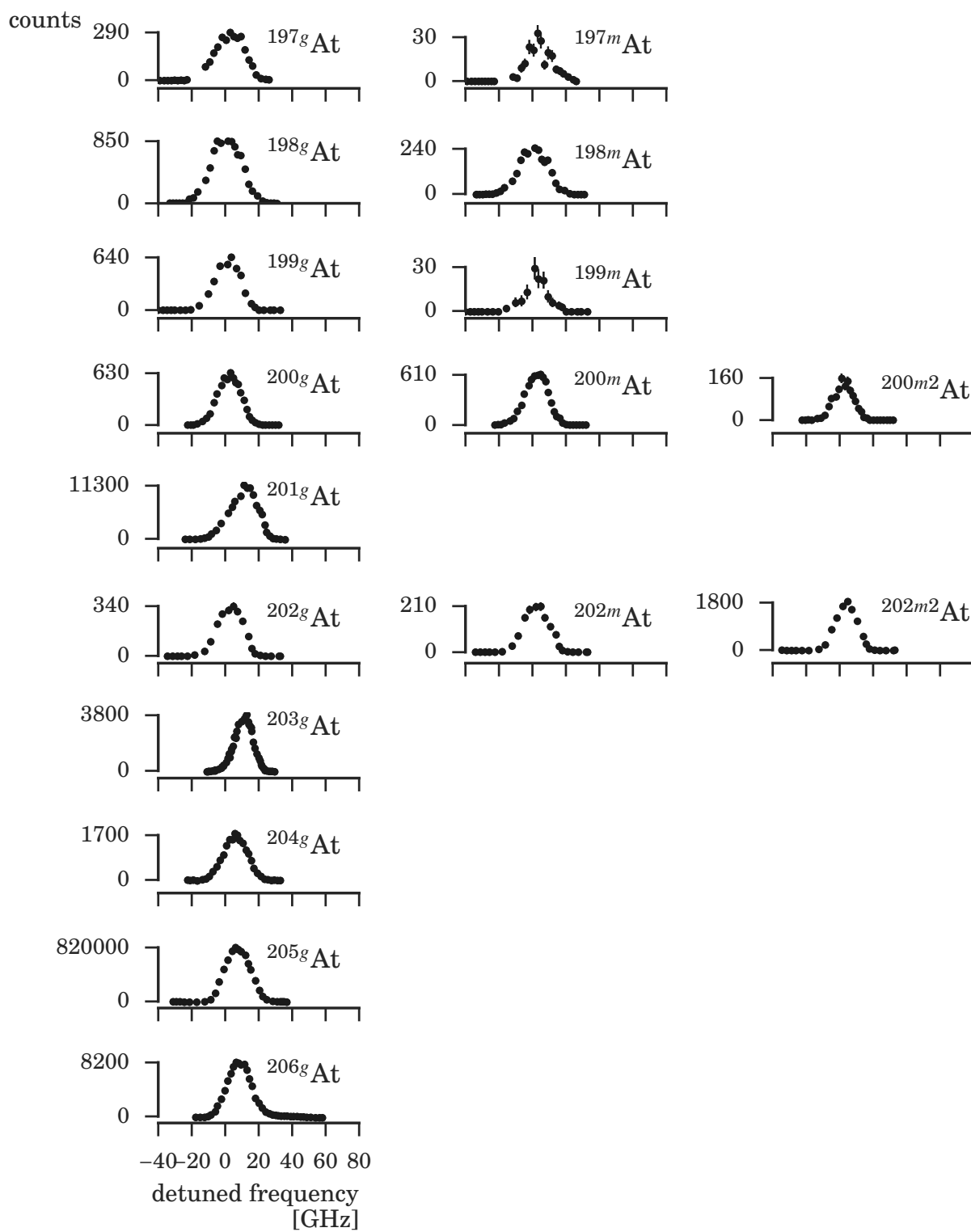
7.2 Isotope shift in astatine

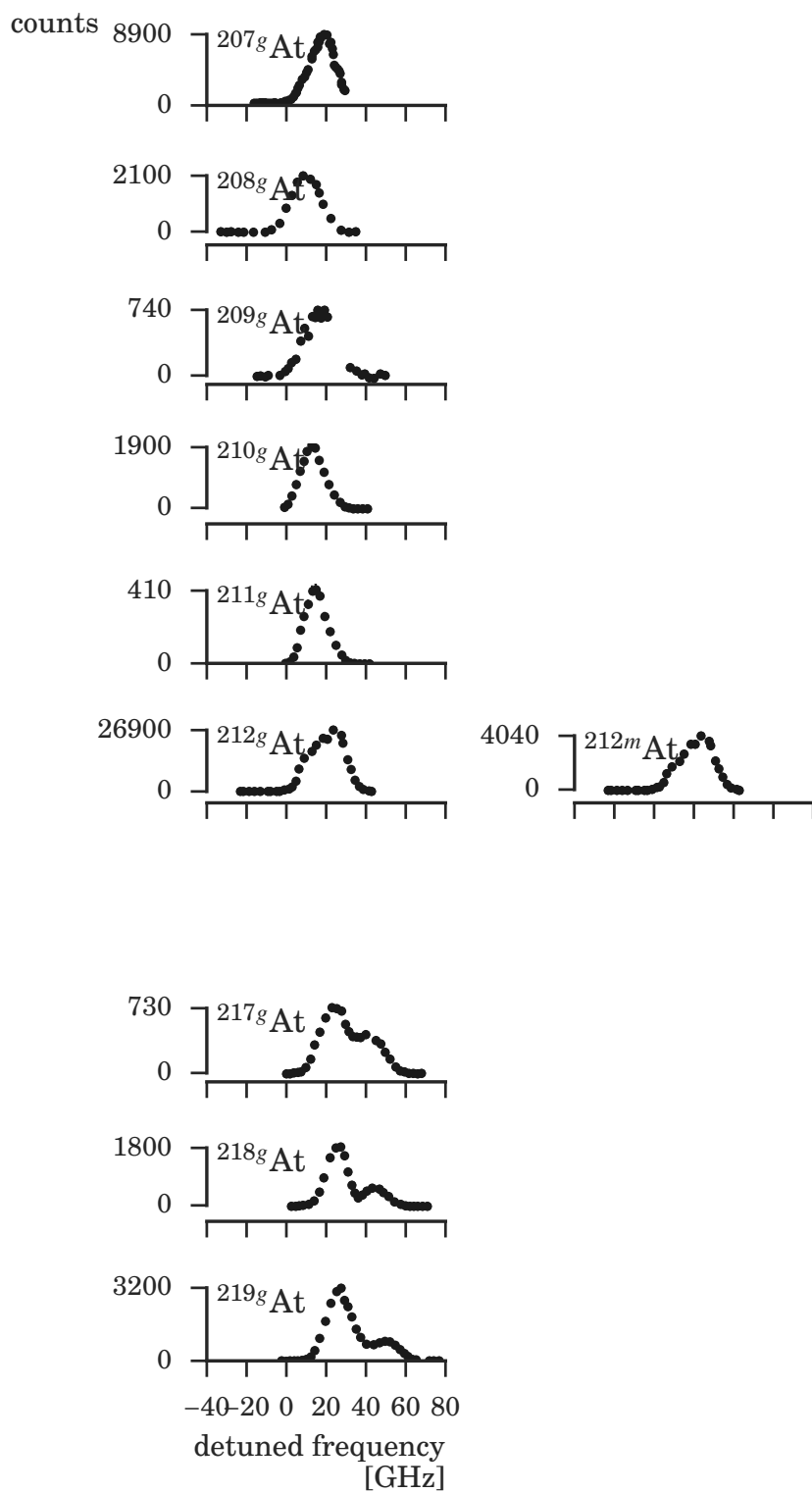
The spectroscopy conducted at TRIUMF and CERN was performed mainly on the neutron-deficient isotopes ^{199}At and ^{205}At , respectively, due to low isobaric contamination at these masses, resulting in a suitable signal-to-noise ratio for ion detection. However, the isotopes requested for experiments at ISAC are $^{221,223}\text{At}$ or heavier if available, i.e., about 20 u away from those isotopes where the atomic

transitions are known. Even for elements with small ISs, the change in electron transition energy caused by this mass difference would be noticeable in laser ion source operations. Particularly heavy elements such as astatine are expected to have a rather large IS. A frequency shift in the FES of approximately 50 GHz was measured for the isotopes ^{199}At and ^{219}At .

At the time when the experiment was scheduled, the heaviest isotope ionized at TRIUMF was ^{217}At with a different excitation scheme, so that the wavelengths for the requested isotope were unknown. For that reason, the FES was scanned for selected isotopes to estimate the IS for the neutron-rich astatine isotopes. The detectors at the ISAC Yield Station are not efficient enough to identify astatine isotopes heavier than ^{220}At which mainly β decays and where γ lines are mostly unknown. Therefore, the RIB was sent to the more sensitive detector of the experiment without characterization. However, by the end of the run none of the requested isotope had been detected. For that reason, every now and then 30 min were provided to the laser ion source group to collect more data for the systematic study of the lighter astatine isotopes. The majority of the scans shown in Figure 7.3 and Figure 7.4 were compiled that way. The scans for the missing isotopes $^{203,207,209}\text{At}$ were taken later with the ThO-target.

Especially noticeable in the scans of the FES is the double peak structure shown in the neutron-rich isotopes $^{217-219}\text{At}$ whose splitting increases with increasing mass. At first it was thought that this phenomenon was caused by the underlying HFS, which could have been a sign of strong deformations in those isotopes. On the other hand it was questionable how, with the known nuclear and atomic spins, a spectrum with only two distinguishable peaks could be achieved. For the evaluation of the ^{212}At spectrum, one of few isotopes where more than one data set was available, it was furthermore seen that the centroid frequency differed by up to 7.5 GHz and the asymmetry of the lineshape varied between the measurements.

Figure 7.3: Scan of the first excitation step in $^{197-206}\text{At}$.

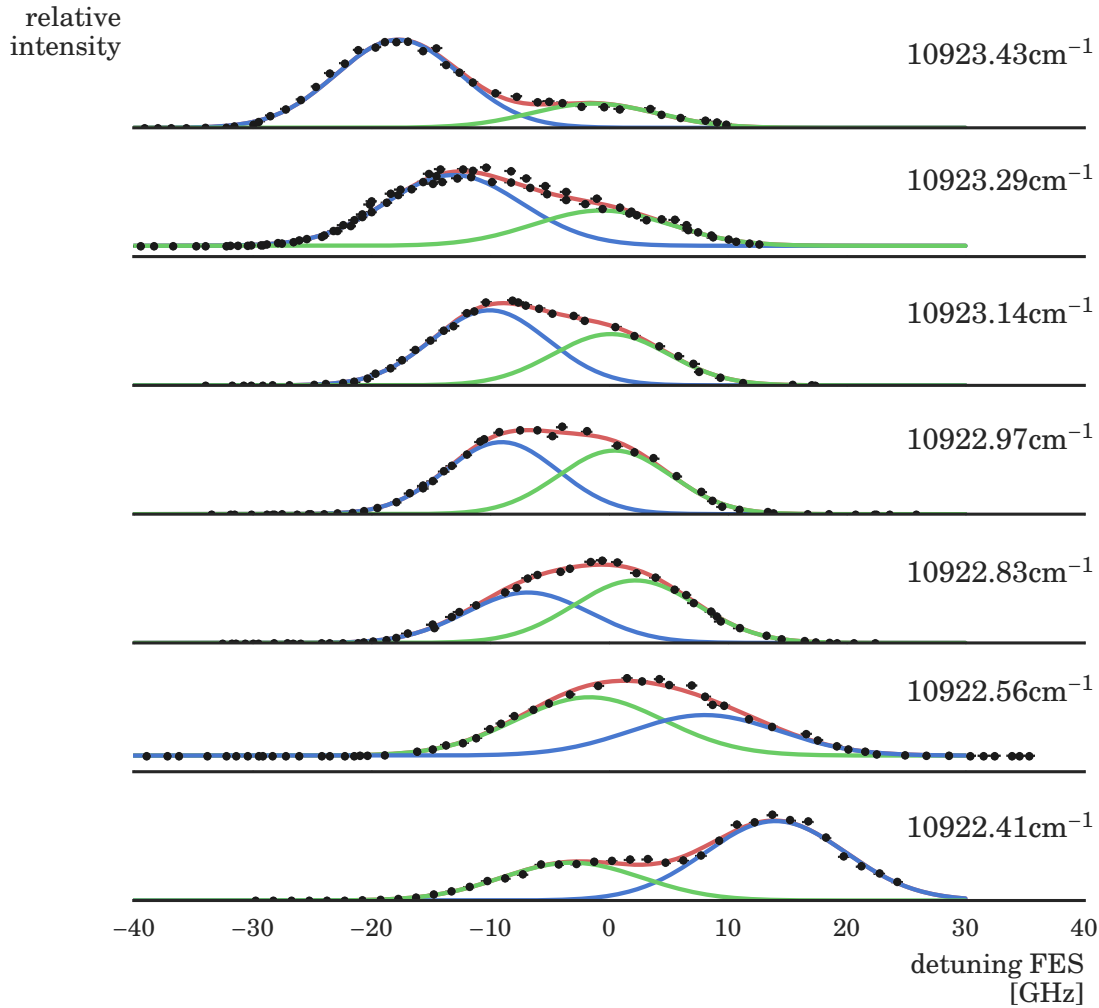
Figure 7.4: Scan of the first excitation step in $^{207-212, 217-219}\text{At}$.

7.2.1 Second excitation step spectroscopy

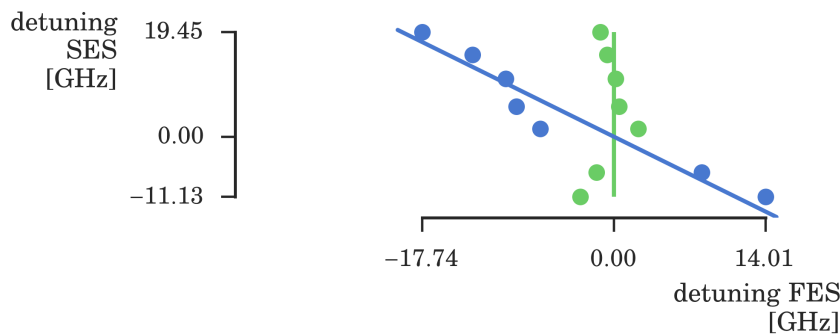
Some of the available time with the ThO-target was used to investigate the observations made in the spectra of the neutron-rich isotopes $^{217-219}\text{At}$ in more detail. Already during the evaluation of the ^{212}At spectra it was seen that the only difference recorded between the datasets was a small shift in the SES of 4.5 GHz. Since the SES has, due to Doppler and saturation broadening, a FWHM of 15 GHz [96], it was not expected that such a small shift compared to the linewidth could have had an effect on the FES.

Nörtershäuser et al. [41, 42] showed however, that in a multistep excitation scheme the lineshape of the scanned transition can be influenced if at least one of the other transitions is detuned. Astatine uses an excitation scheme with two resonant transitions as shown in Figure 7.1, so if the SES is not on resonance when the FES is scanned, a double peak spectrum can be observed. In such a spectrum, one of the peaks coincides with the resonance frequency of the FES. The dominant peak on the other side arises if the total photon energy of both lasers matches the resonance energy of the second excited state. This effect is especially prominent if the transitions are well saturated, as was the case for the scans shown in Figures 7.3 and 7.4, which were conducted with the full laser power available.

In order to verify whether a detuned SES could cause for the double peak spectra of $^{217-219}\text{At}$, FES-scans were performed systematically at ^{217}At for different wavelengths of the SES. It can be clearly seen in Figure 7.5a, that the dominant peak, moves from lower to higher frequencies, with decreasing frequency from the SES-laser. In order to determine the resonance frequencies for each excitation step, every scan was fitted with two Gaussians of identical linewidth. The centroid frequency of each Gaussian can be assigned as either matching the photon energy of the first excited state (in Figure 7.5 indicated in green), or, in combination with the second laser, the total photon energy of the second excited state (in Figure 7.5 indicated in blue). At first the resonance frequency of the FES can be determined from the weighted mean of the frequencies involved in this transition.



(a) FES scans at different SES wavelengths. The fixed wavelength for the SES is indicated on the left.



(b) Centroid frequency of the two Gaussians with respect to the wavelength of the SES.

Figure 7.5: Wavelength determination for FES and SES. The FES is scanned at different fixed SES wavelengths. The resulting double peak structure is fitted with two Gaussians, where the dominant peak (indicated in blue) corresponds to the wavelength where the sum of the photon energy of the two lasers matches the level energy of the second excited state. The peak indicated in green belongs to the resonance frequency of the first excited peak.

The FES for ^{217}At can in this way be assigned to $46\,235.09(2)\text{ cm}^{-1}$. The frequency of the dominant peak in dependency on the detuned SES forms a line with a slope of $-0.99(14)$. The resonance frequency of the SES corresponds now to the intersection between these two lines, which occurs at a wavelength of $10\,922.78(13)\text{ cm}^{-1}$.

The field shift, which is the dominant contribution to the IS of heavy elements, is proportional to the change of the electron density at the nucleus. With increasing principle quantum number n the electron density at the nucleus decreases; therefore, in most cases the IS for higher excited states can be neglected. Even though the electron configuration for the $57\,157.1\text{ cm}^{-1}$ state is unknown, considering that the first step is a $6p \rightarrow 7s$ transition, the excited step must be a $7s \rightarrow np$ transition, which however again implies a large IS [49]. Therefore the double peak spectrum of $^{217-219}\text{At}$ suggests an underrated IS of the SES.

For a better understanding of this IS, the SES was scanned for selected isotopes. Since at this point neither the first nor the second step are exactly known, the laser power was reduced to $P_{\text{FES}} = 2.7\text{ mW}$ and $P_{\text{SES}} = 40\text{ mW}$ to avoid any artificial shifts or structure. In order to determine the CoG of each scan, a similar technique was used as described for silver in Section 5.4.

Each scan of the $I = 9/2$ isotopes $^{199,205,217}\text{At}$ in Figure 7.7 shows four distinguishable peaks, therefore it was assumed that the HFS of the $^4\text{P}_{3/2}$ -state is larger than that of the $J = 5/2$ -state. At first each scan, except the one for the ^{212g}At isotope, was fitted with four Gaussians of equal linewidth. Now each peak can be assigned to one component of the $^4\text{P}_{3/2}$ HFS as shown in Figure 7.6. Since in this spectrum the larger HFS splitting is assumed to be in the lower excited state, similar to silver, the peak with the highest frequency is assigned to the lowest total angular momentum and vice versa. Every peak contains three underlying HFS components whose theoretical relative intensities can be calculated by using eq. (3.31). Again, the CoG was defined by the weighted mean of the four peak centroids where the relative intensities serve as weighting factors. The result of this evaluation is shown in Figure 7.7. Since for the ^{212g}At isotope with $I = 1$ no HFS can be resolved, the CoG was assigned by fitting a single Gaussian to this scan. The $I = 9$ isomer of this

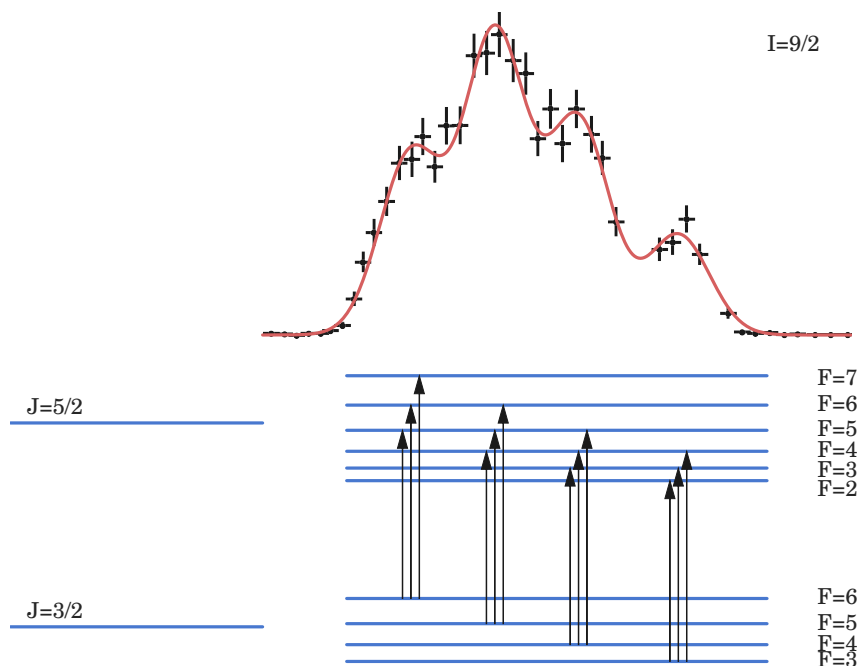


Figure 7.6: HFS of the SES in astatine in an $I = 9/2$ isotope.

isotope was treated the same way as the $I = 9/2$ isotopes, i.e. by fitting four separated Gaussians to that scan and assigning the corresponding Racah intensities to each Gaussian. The noticeable asymmetric shape of this peak is caused by the fact that the FES was adjusted for a scan of the ^{212}gAt isotope. Therefore in order to determine the real CoG, the FES isomer shift of 1.8 GHz had to be added to the result from the weighted mean calculation. Furthermore, it can be seen that for this particular isotope it is possible to separate the ground state from the isomer if the laser power of the SES is reduced to close to the saturation intensity.

Since the SES shows a pronounced HFS, it was also tested whether the A - and B -factors for the $^4\text{P}_{3/2}$ state could be identified. Since the evaluation used for actinium, section 6.2, only provides meaningful results if the correct starting values for the fit are chosen, this technique cannot be used for astatine where so far no literature values for the HFS are available. Based on the results of the fit for the CoG and the assumption that the splitting of the $J = 5/2$ state can be neglected, the A -

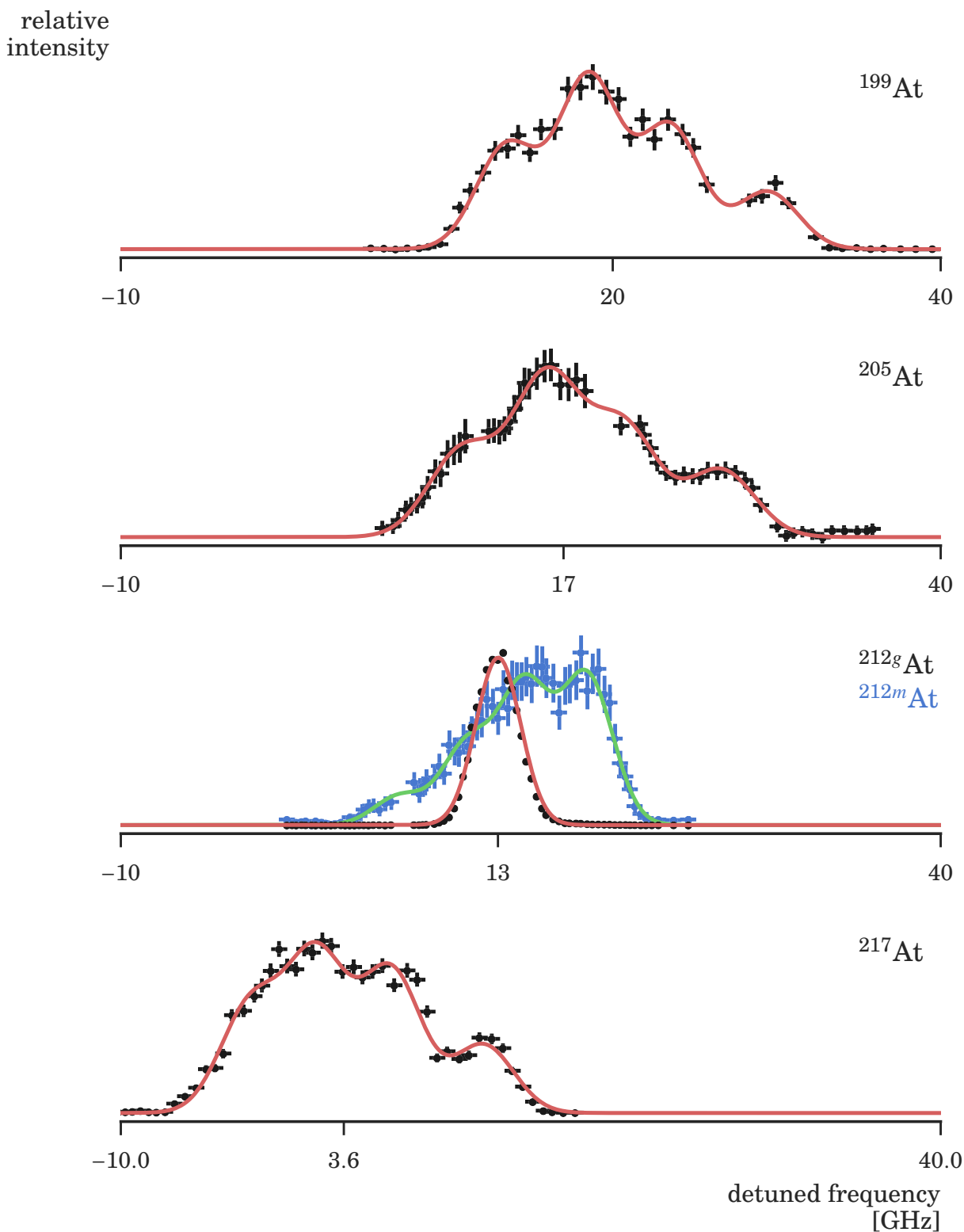


Figure 7.7: Scans of the second excited states in astatine for selected isotopes. The frequency is relative to the previously determined wavelength of ^{217}At of $10\,922.78(13)\,\text{cm}^{-1}$. The CoG for the ground state of each isotope is indicated.

Table 7.1: Isotope shift and hyperfine structure constants for the second excitation step in astatine.

isotope	$\delta\nu_{IS}^{A,217}$ [GHz]	A [GHz]	B [GHz]
199	16.3(3)	1.01(1)	-2.4(4)
205	13.4(6)	1.04(5)	-2.1(4)
212g	9.4(3)		
212m	10.5(5)	0.39(4)	-0.6(2)
217	0	0.90(2)	-2.2(4)

and B - factors were calculated by solving a linear equation system of the form

$$\text{CoG} - \nu_i = A \frac{K_i}{2} + B \frac{\frac{3}{4} K_i (K_i + 1) - I(I + 1) J(J + 1)}{2(I - 1)(2J - 1)IJ}, \quad (7.1)$$

where total angular momentum F_i contained in the coupling constant K_i by eq. (3.34) is assigned to the center frequency ν_i in the same manner as for calculating the CoG. The results of this calculation are shown in Table 7.1.

In order to demonstrate how reliable these results are, the same procedure was tested on the ^{227}Ac scans from Figure 6.7, where literature values are available [88]. Since for actinium only the HFS of the excited state can be resolved, the assignment of the total angular momentum F to the individual peaks has to be reversed compared to astatine. Therefore, higher frequencies belong to higher total angular momenta. Furthermore, the sign in the y-matrix in eq. (7.1) has to be inverted. From the results in Table 7.2, it can be seen that the A -factor agrees reasonably well with the literature value, but not the B -factor, in magnitude nor sign. Considering however that for actinium the B -factor for the ground state is with $B = 595.6(21)$ MHz larger than for the excited state, the result for the B -factor is also acceptable, since the ground state contributes to this calculation with the opposite sign. Hence, the evaluation of the HFS by solving the linear equation system with the assumptions made above allows one to determine the magnitude of the A - and B -factor involved, but not to which state the constants belong.

Table 7.2: The method used for astatine to calculate the HFS constants A and B , applied to ^{227}Ac and compared with the known literature values. Despite the small uncertainties for the calculated A -factor, the result comes close to the literature value. The B -factor however disagrees in magnitude and sign. The main reason for the disagreement is that the B -factor for the ground state is larger than that of the excited state.

	A [MHz]	B [MHz]
literature	2104.8(5)	110.1(22)
lin. eq. system	1976(19)	-744(54)

7.2.2 First excitation step spectroscopy

From the number of distinguishable HFS components in the SES, it is assumed that the main contribution to the HFS arises from the common $^4\text{P}_{3/2}$ state. Therefore in the FES a similar HFS splitting is expected as in the SES. However, even with reduced laser power it was not possible to resolve the underlying HFS in the FES as shown in the example of ^{212}At (Figure 7.8). The main difference between these two transitions are the wavelengths. Since the Doppler broadening scales linearly with the photon energy as shown eq. (3.15), a larger broadening for the FES is expected. For ^{212}At , the Doppler broadening at a temperature $T = 2000\text{ K}$ is calculated to be $\Delta\nu_{D,\text{FES}} = 3.1\text{ GHz}$ and $\Delta\nu_{D,\text{SES}} = 0.7\text{ GHz}$. Furthermore, the wavelength of the FES is generated by quadrupling the fundamental wavelength of 865 nm of a Ti:Sa-laser which doubles laser linewidth. Finally, even though this fact can be neglected for in-source laser spectroscopy, commonly higher excitation steps have weaker transition probabilities compared to ground state transitions so that the natural linewidth of the SES is also one to two orders of magnitudes narrower. The difference can be seen from the scans of the ^{212g}At isotope in Figures 7.7 and 7.8, which do not show a HFS splitting in either case. For this isotope, the linewidth was fitted to $\text{FWHM}_{\text{SES}} = 1.6(1)\text{ GHz}$ compared to the $\text{FWHM}_{\text{FES}} = 3.9(1)\text{ GHz}$.

The majority of scans for the FES shown in Figures 7.3 and 7.4 were performed with the full laser power available. Due to the large Doppler broadening, each scan except those for the neutron-rich isotopes was fitted with a single Gaussian. As discussed in section 7.2.1, it is possible to

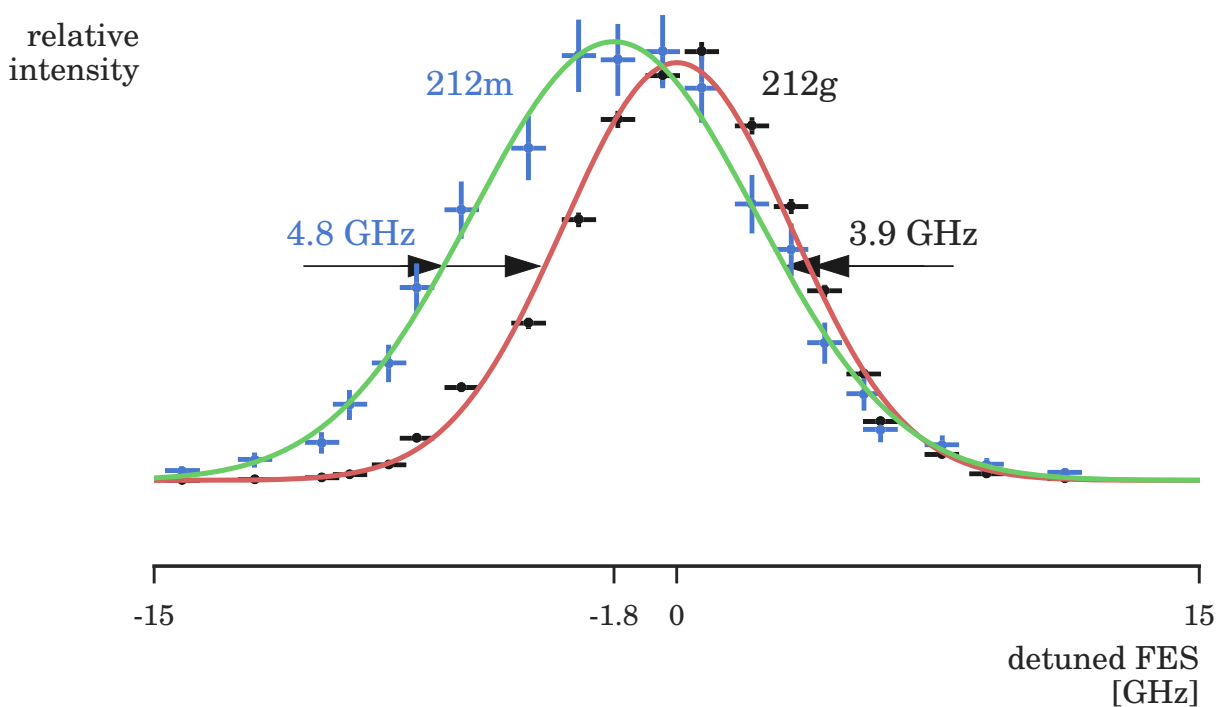


Figure 7.8: Scan of the FES of ^{212}At with reduced laser power. The laser power was reduced to below the saturation powers of the transitions of $P_{\text{FES}} = 1 \text{ mW}$ and $P_{\text{SES}} = 20 \text{ mW}$. Even though the transition for the isomer is $0.9(2) \text{ GHz}$ wider than that of the ground state and is shifted by $-1.8(3) \text{ GHz}$, the difference is too small for isomer separation.

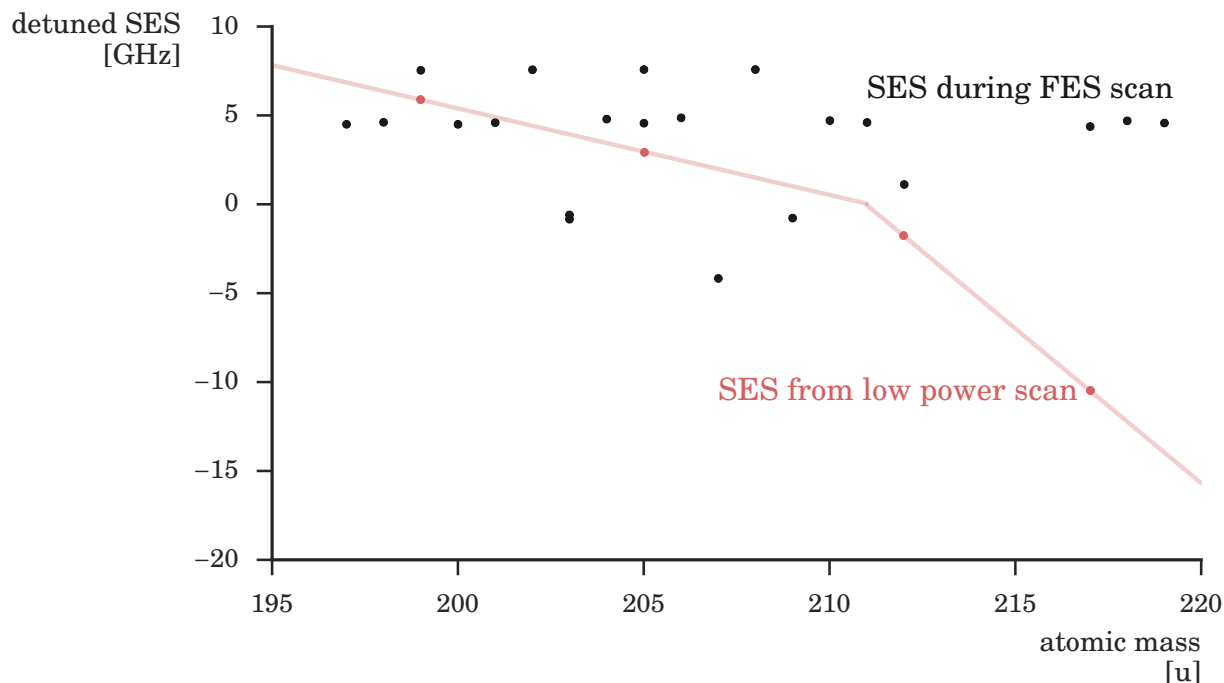


Figure 7.9: Isotope shift of the SES (red) compared to the wavelength where the SES was during the FES scan (black).

introduce shifts and splittings if the SES is off-resonance during the FES scan. This was the case for most of the scans since the IS of the SES was underestimated during the FES scans. In order to compensate for these shifts, it was first investigated how much the SES was detuned. Therefore, the IS listed in Table 7.1 were used as an aid. To first approximation it is assumed that the IS behaves linearly towards as well as beyond the neutron-shell closure at $A = 211$, as it has been shown for the neighboring chain of isotopes as shown in Figure 1.1. Consequently, the IS of the SES for neutron-deficient isotopes has a slope of $m = -0.49(17) \text{ GHz u}^{-1}$ and the neutron-rich of $m = -1.74(34) \text{ GHz u}^{-1}$. The SES was in average 3 GHz detuned from the resonance frequency as shown in Figure 7.9.

By using the results from the scans shown in Figure 7.5, it is possible to estimate the IS of the FES. From the example of ^{212}At it can be seen that the isomer shift cannot be ignored. Since not enough information is available for each isomer, it was assumed that the shift from the detuned SES is larger than the isomer shift, and was therefore treated like the IS of the nuclear ground state.

Table 7.3: Isotope shift for first excitation step in astatine corrected for the detuned second excitation step.

isotope	$\delta\nu_{\text{IS}}^{A,211}$ [GHz]	isotope	$\delta\nu_{\text{IS}}^{A,211}$ [GHz]	isotope	$\delta\nu_{\text{IS}}^{A,211}$ [GHz]
197g	-19.0(11)	201	-14.7(10)	208	-3.1(13)
197m	-18.9(13)	202g	-14.3(13)	209	-3.1(10)
198g	-20.9(10)	202m1	-13.0(13)	210	-2.5(12)
198m	-21.3(10)	202m2	-12.0(11)	211	0
199g	-17.1(10)	203	-12.8(13)	212g	4.0(8)
199m	-16.0(10)	204	-12.5(10)	212m	2.2(11)
200g	-18.5(9)	205	-9.8(10)	217	22.6(10)
200m1	-18.5(9)	206	-8.6(9)	218	25.3(10)
200m2	-18.7(10)	207	-8.2(11)	219	29.2(10)

The IS for the neutron-rich isotopes was determined directly from the center frequency of the low intensity side peak in the shown double-peak structure. The results of these calculations are listed in Table 7.3.

7.2.3 King plot

Since the majority of isotopes shifts were not directly measured, but deduced by introducing a correction factor to compensate for detuned steps in the resonant excitation ladder, it is of interest to test the consistency of the determined ISs. The consistency can be evaluated with the King plot analysis (more details in section 3.4.2), where the IS of two different transitions of the same element are plotted against each other. By multiplying the ISs with a modification factor $\frac{m_A m_{A'}}{m_A - m_{A'}}$, the isotope-dependent parameters cancel each other, and the resulting modified isotope shift should lie on a straight line. Such a King plot for the transitions of the first and second excitation step is shown in Figure 7.10, where ^{217}At was used as the reference isotope. Considering how many assumptions have been made during the evaluation of the isotope shifts, the result of the King plot is surprisingly good, which is an indication, but not proof of the quality of the data.

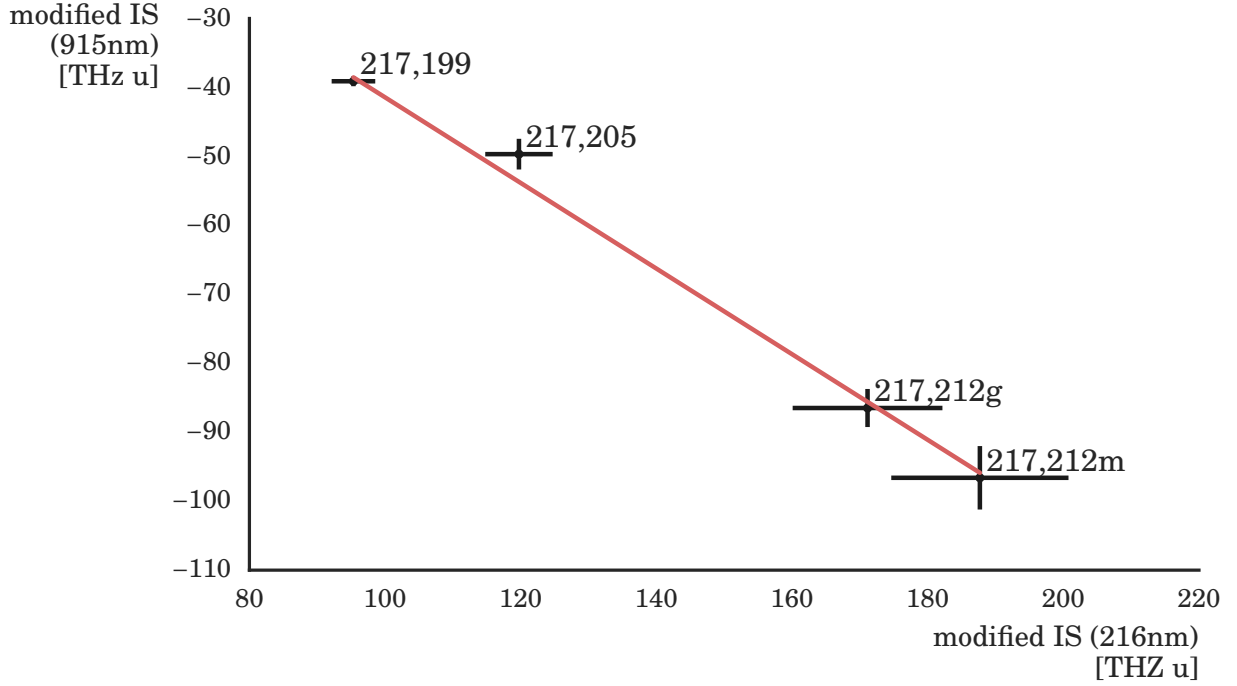


Figure 7.10: King plot of the FES transition versus the SES transition in astatine.

Originally, the King plot analysis was used to determine the field shift F - and mass shift M -parameters if they were known for one transition but not for the other. For astatine the field shift constant F has been provided by theory using the coupled cluster method for the two ground state transitions. The field shift constant for the transition to the $^4P_{5/2}$ state was calculated to $F = 32.97 \text{ GHz fm}^{-2}$ and the transition to the $^4P_{3/2}$ state which was used for the spectroscopy here discussed was calculated to $F = 30.39 \text{ GHz fm}^{-2}$ [98]. Using this result, the field shift constant for the SES can also be calculated. Therefore the slope of the fit in Figure 7.10 is multiplied with the field shift factor of the reference transition, hence $F_{915 \text{ nm}} = -18.6(11) \text{ GHz fm}^{-2}$.

Since these values were not available during the evaluation process of this thesis, the F parameter had been previously estimated by using a technique related to the King plot analysis [10]. The field shift is proportional to the change of the electron density at the nucleus $\Delta |\Psi(0)|^2$. By assuming that $\Delta |\Psi(0)|^2$ is the same for all $6p^n \rightarrow 7s$ transitions, it is possible to compare ISs for these transitions in different elements and extract the F -parameter from there, as has been demonstrated previously on bismuth [99]. In order to compare the ISs, they first have to be normalized to a common isotone

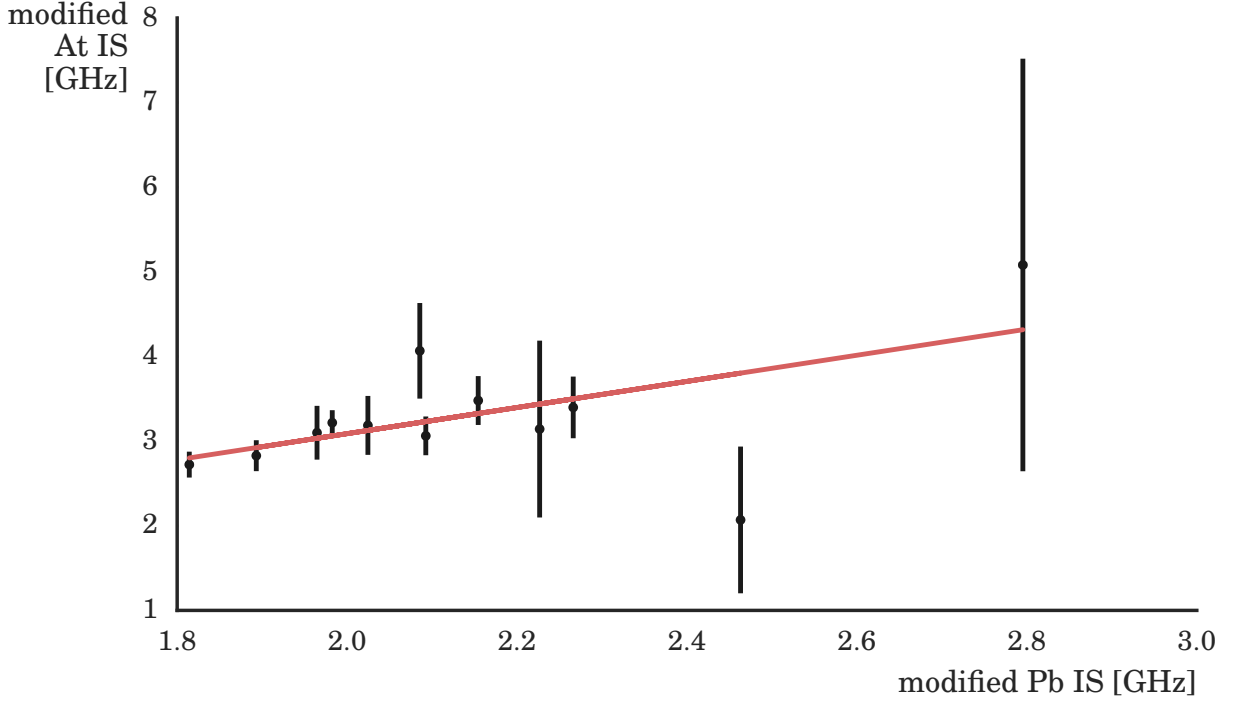


Figure 7.11: Modified King plot of the lead against the astatine isotope shift.

pair, which was chosen to be $N, N' = 124, 126$, and results in a modification factor μ of

$$\mu = \frac{2(Z + N)}{(Z + 124)(N - 126)}. \quad (7.2)$$

It is expected that these modified ISs also results in a straight line when plotted against each other, where the intersection is related to M and the slope to F . Such a modified King plot for the lead against the astatine isotopes is shown in Figure 7.11. Since the mass shift is expected to be negligible, the line is forced to go through the origin. From the resulting slope m of the line, it is possible to extract the field shift of the ground state transitions in astatine to

$$F_{\text{At}(216 \text{ nm})} = F_{\text{ref}} \cdot m. \quad (7.3)$$

This method assumes that the mean squared charge radius changes uniformly for all considered isotope chains. Only neutron-deficient isotopes $112 \leq N \leq 126$ have been used for this comparison,

Table 7.4: Field shift parameters for the ground state transition in astatine, determined from the modified King plot of Tl, Pb, Bi and Po.

isotopes	transition [nm]	F_{ref} [GHz fm ⁻²]	$F_{\text{At}(216\text{ nm})}$ [GHz fm ⁻²]	references
183–207Tl	535.2	20.79	37.0(23)	[100, 101, 102, 103]
196–208Pb	283.3	20.26(18)	31.8(10)	[104, 105]
202–209Bi	306.8	27(3)	32.2(54)	[99, 106, 107]
200,202,204–210Po	255.8	28.383	28.6(28)	[108, 13]

since for those isotopes, nuclei with a spherical shape are expected. This procedure has been applied to four different isotope chains of the elements thallium, lead, bismuth, and polonium, and the results are listed in Table 7.4. Except for the result from the thallium comparison, all field shift parameters are in good agreement with each other. A possible reason for the discrepancy is that, even though it is a $6p \rightarrow 7s$ transition, it does not start from the atom ground state, but from a lower excited state at 7792.7 cm^{-1} . Therefore, this result was excluded from the further treatment. Consequently the field shift parameter for the first excitation step in astatine can be determined to $F_{216\text{ nm}} = 31.4(19) \text{ GHz fm}^{-2}$. This result agrees well with the value provided by theory.

7.3 Nuclear charge radius

Also for astatine, the change of the mean squared charge radius $\delta \langle r^2 \rangle$ can be determined by using eq. (6.2), assuming that the effect of the mass shift and the Seltzer moments can be neglected. As described above, the ISs of the two resonant transitions strongly depend on each other. For the FES the IS was estimated by considering a linear trend for the IS of the SES. However as shown for the example of ^{212}At , the isomer shift may vary by a few GHz from that of the nuclear ground state, and therefore would not have a linear behavior. Since ^{212}At is the only isotope with an isomer where the frequency shift has been measured in both transitions, it is only possible to determine the $\delta \langle r^2 \rangle$ reliably for the nuclear ground state. The deduced $\delta \langle r^2 \rangle$ using the isotope shift for the FES listed in Table 7.3 and a field shift constant of $F_{216\text{ nm}} = 30.39 \text{ GHz fm}^2$ are shown in Table 7.5.

Table 7.5: Change of the mean squared charge radii of astatine derived from the optical isotope shift measurement of the first excitation step.

A	$\delta \langle r^2 \rangle^{A,211}$ [fm ²]	A	$\delta \langle r^2 \rangle^{A,211}$ [fm ²]	A	$\delta \langle r^2 \rangle^{A,211}$ [fm ²]	A	$\delta \langle r^2 \rangle^{A,211}$ [fm ²]
197	-0.62(3)	202	-0.47(4)	207	-0.27(4)	212	0.13(3)
198	-0.69(3)	203	-0.42(4)	208	-0.10(4)	217	0.74(3)
199	-0.57(3)	204	-0.41(3)	209	-0.10(3)	218	0.83(3)
200	-0.61(3)	205	-0.32(4)	210	-0.08(4)	219	0.96(3)
201	-0.48(3)	206	-0.28(3)	211	0		

It should be noted that there seems to be a measurement error for the IS of ^{208}At ($N = 123$) which shows a larger mean squared charge radius than expected. The IS of this isotope was also already an outlier in all modified King plots used to determine the field shift constant, where the example of lead is shown Figure 7.11. The measurement for the isotope shift of this isotope was the only one where the wavelength of the SES was not simultaneously monitored. Even though the wavelength was recorded when it was last measured, which was shortly before the measurement, it is possible that the SES drifted during the measurement. For that reason an uncertainty for the wavelength of the SES was added to calculate the isotope shift of the FES.

The nuclei of the lead isotope chain are in this mass region, and due to their closed proton shell ($Z = 82$), they are expected to be spherical. For that reason, $\delta \langle r^2 \rangle_{\text{At}}$ is in Figure 7.12 compared with $\delta \langle r^2 \rangle_{\text{Pb}}$ [11].

Since the neutron-deficient lead isotopes show an increase of deformation at $N = 115$ [109], in addition also the gradients for deformed nuclei are indicated in this graph, where the $\delta \langle r^2 \rangle$ for a spherical nucleus is described by the droplet model [110].

In the liquid drop model, it is assumed the protons and neutrons are uniformly distributed within a spherical nucleus. Therefore the volume is proportional to the number nucleons A . This model describes the change of the nuclear charge radius well for nuclei close to stability, but fails for a long chain of isotopes of the same element. For that reason the droplet model [110] has been introduced, which takes the different distribution between proton and neutrons into account. For a

spherical nucleus, the charge radius in droplet model is expressed as:

$$R = R_0 A^{1/3} (1 + \epsilon), \quad (7.4)$$

where

$$\epsilon = \frac{1}{K} \left(-a_2 A^{-1/3} + L\delta^2 + c_1 Z^2 A^{-4/3} \right), \quad (7.5)$$

$$\delta = \frac{I + \frac{3}{16} (c_1/Q) Z A^{-2/3}}{1 + \frac{9}{4} (J/Q) A^{-1/3}}, \quad (7.6)$$

$$I = \frac{N - Z}{A}. \quad (7.7)$$

The coefficients used in this model were optimized for the double magic nucleus ^{208}Pb to

$R_0 = 1.16$ fm, the nuclear radius constant,

$c_1 = \frac{3}{5} \frac{e^2}{R_0} = 0.7322$ MeV, the Coulomb energy coefficient,

$a_2 = 23$ MeV, the surface energy coefficient,

$J = 29.5$ MeV, the symmetry energy coefficient,

$Q = 45$ MeV, the effective surface stiffness,

$K = 240$ MeV, the compressibility coefficient, and,

$L = -35$ MeV, the density symmetry coefficient [111].

Although a more precise version the droplet model [110] is able to describe deformed nuclei, the gradients shown in Figure 7.12 are generated by using eq. (3.51).

It can be seen that for astatine the nuclei close to the neutron shell closure also show a spherical behavior. Due to the large uncertainties and limited data, however, it is not obvious whether astatine follows the trend of lead, which stays relatively spherical, or if like polonium it shows an early onset

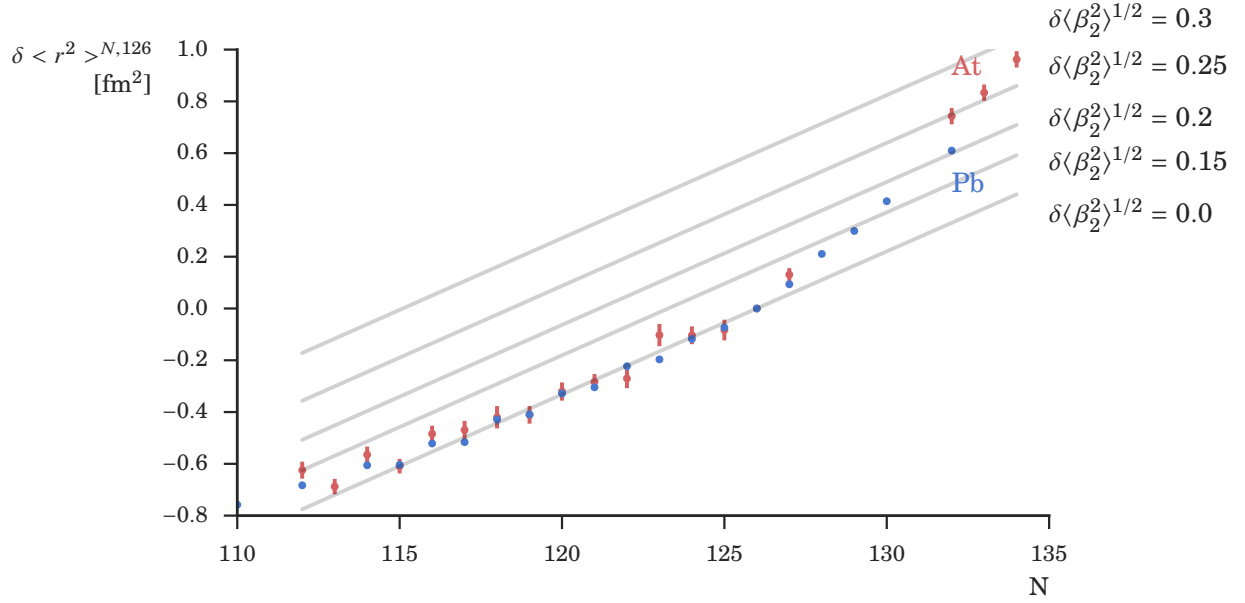


Figure 7.12: Comparison of the means squared charge radius between astatine and lead. The prediction from the droplet model for different deformation parameters $\delta \langle \beta_2^2 \rangle^{1/2}$ is included in the plot.

on deformation [13]. Obvious is that astatine in the mass region around $N = 115$ displays a more distinct odd-even staggering. In reality the odd-even staggering is expected to be larger than shown in Figure 7.12, since for the correction of the SES a linear behavior was assumed. Furthermore it can be seen that the changes of the mean squared charge radii in the neutron-rich astatine isotopes exceed those seen in lead.

7.4 Astatine spectroscopy summary

The IS for the FES of $^{197-212,217-219}\text{At}$ has been deduced to be $1.54(4) \text{ GHz u}^{-1}$ for $^{197-211}\text{At}$ and $3.67(4) \text{ GHz u}^{-1}$ for $^{211-219}\text{At}$. This shows that the lineshape and therefore the CoG of the FES strongly depends on correct setting of the SES. For that reason also the isotope shift of the SES has been measured for $^{199,205,212,217}\text{At}$, which resulted in $-0.49(17) \text{ GHz u}^{-1}$ for neutron-deficient isotopes, and $-1.74(34) \text{ GHz u}^{-1}$ for isotope beyond the neutron shell closure. With this information available it was possible to correct the measured CoG of the FES for the detuned SES.

By comparing the $6p \rightarrow 7s$ transition ISs of neighboring elements where the field-shift factor F is known, it was possible to also estimate the field-shift factor for astatine to $F = 31.4(19) \text{ GHz fm}^2$. This agrees well with the theoretical calculation of $F = 30.39 \text{ GHz fm}^2$. Furthermore the field shift factor for the SES was calculated using the King plot-analysis to $F = -18.6(11) \text{ GHz fm}^{-2}$. Finally the change of the mean squared charge radius $\delta \langle r^2 \rangle$ was determined and compared with the nuclear droplet model.

8 | Summary

In this work, in-source resonant ionization laser spectroscopy has been performed on isotopes of three different elements: silver, actinium and astatine. The main purpose for including the **silver** data in this thesis was to demonstrate the evaluation process and advantages of monitoring the radioactive decay product at the ISAC Yield Station over ion counting on a CEM or FC. The laser frequency scans were analyzed by evaluating the β decay of $^{114,115}\text{Ag}$ as well as the γ decay of $^{116-119}\text{Ag}$. Nuclei that emit α particles are treated the same way as those emitting γ rays. The main advantages of this detection method are

- the possibility to detect laser ionized species despite high isobaric background, provided that the isotope of interest and the background isotope have different decay properties,
- the separation of spectra generated from nuclei in the ground or isomeric state,
- assignment of α - or γ -energies to specific isotope and isomers of interest.

It had been the first time that RADRIS was applied at TRIUMF. Therefore there are still opportunities to improve the method currently applied at TRIUMF.

- An upgrade of the motor that operates the etalon by a hysteresis-free model with encoder¹ would allow a reliable repeat of the same scan numerous times. At the moment, the only possibility to increase the count rate for each measurement, is to increase the measurement time at each frequency step. On the other hand, it is not advisable to exceed a scan time

¹e.g. New Focus 8310 Closed Loop Picomotor™ actuator with integrated rotary encoder

of more than two hours, as the scan quality might suffer from possible RIB or proton beam fluctuations. In order to compensate for those fluctuations, it would be better to have a number short scans with low count rate, that could be added together. This integration would greatly benefit from repeatable frequency scans.

- For isotopes that have to be analyzed by evaluating their half-lives, as it was the case for $^{114,115}\text{Ag}$, it would be helpful if the number of decay cycles per frequency step could be varied. With the current scanning program the frequency changes with each decay cycle. Most isotopes are analyzed by measuring the number of counts in the assigned α or γ energies, or the overall number of β particles. In these cases it is possible to increase the count rate by increasing the collection time. However, for short-lived isotopes whose half-life has to be fitted for evaluation, a longer collection time does not lead to a better fitting result, since the signal saturates after a couple of half-lives.
- Furthermore a direct count rate monitor as a function of the laser frequency could be implemented.

It was not possible to extract the IS, change of the mean squared charge radius $\delta \langle r^2 \rangle$, and the nuclear magnetic dipole moment μ for the silver isotope studied here, as no reference isotope was measured, and the nuclear spin assignments are not known for all isotopes. However for all isotopes, except ^{114}Ag , a separation between the ground- and isomeric-state using different excitation frequencies was demonstrated. As a result, isobarically clean beam from these six isotopes can be delivered for experiments. With the determined frequencies fast and easy switching between ground and isomeric states can also be performed without having to rescan the entire spectrum, which adds another capability to the investigation of nuclear isomers and their decay chains.

The **actinium** isotopes were extracted from a previously irradiated target. For this element two different excitation schemes have been tested. With the most promising excitation scheme the IS and HFS has been measured for the neutron-rich $^{225-229}\text{Ac}$ isotopes. The HFS constants A and B were extracted, and the nuclear magnetic dipole moments μ were deduced. Furthermore the

nuclear spin for ^{226}Ac was confirmed. The laser ionization scheme has been used repeatedly now to extract ^{225}Ac for life science experiments.

In combination with the neutron-deficient actinium isotopes $^{212-215}\text{Ac}$ that have been measured at the LISOL-facility in Leuven (Belgium) [90, 112], actinium is the heaviest element so far where the charge radius has been measured across the neutron shell closure $N = 126$. Although the nuclear spin for ^{229}Ac has been measured as $I = 3/2$, the nuclear dipole moment $\mu = 1.7(2) \mu_N$ has been measured to be larger for the lighter isotopes $^{225,227}\text{Ac}$ with the same nuclear spin at $\mu = 1.1(1) \mu_N$ and $1.2(1) \mu_N$ respectively. A similar effect has previously been observed for $^{227,228}\text{Fr}$ where the nuclear ground states are dominated by a proton hole in the $\pi 3s_{1/2}$ orbital [113].

For **astatine** the modification of the lineshape for the FES as a result of a detuned SES was discussed. For that purpose the IS of the SES was measured for $^{199,205,212,217}\text{At}$. It has been shown that with laser power below saturation the ground and isomeric state in ^{212}At can be ionized separately. Knowing the trend of the SES IS, the shifts in the CoG for the FES scan can be corrected, and the IS for the FES was thus extracted. In order to verify the consistency of the ISs, a King plot analysis between the first and second excitation step IS was performed. Now that the trends of the IS for the FES and SES are known, the search for neutron-rich astatine isotopes for experiments can resume. It is expected that the yields for the so far heaviest detected isotope ^{219}At can be increased by one order of magnitude.

An overview of the changes of the mean squared charge radii $\delta \langle r^2 \rangle$ extracted from isotope shift measurements for nuclei around the proton shell closure $Z = 82$ (indicated in blue) are shown in Figure 8.1. The complementing results from this work are indicated in red.

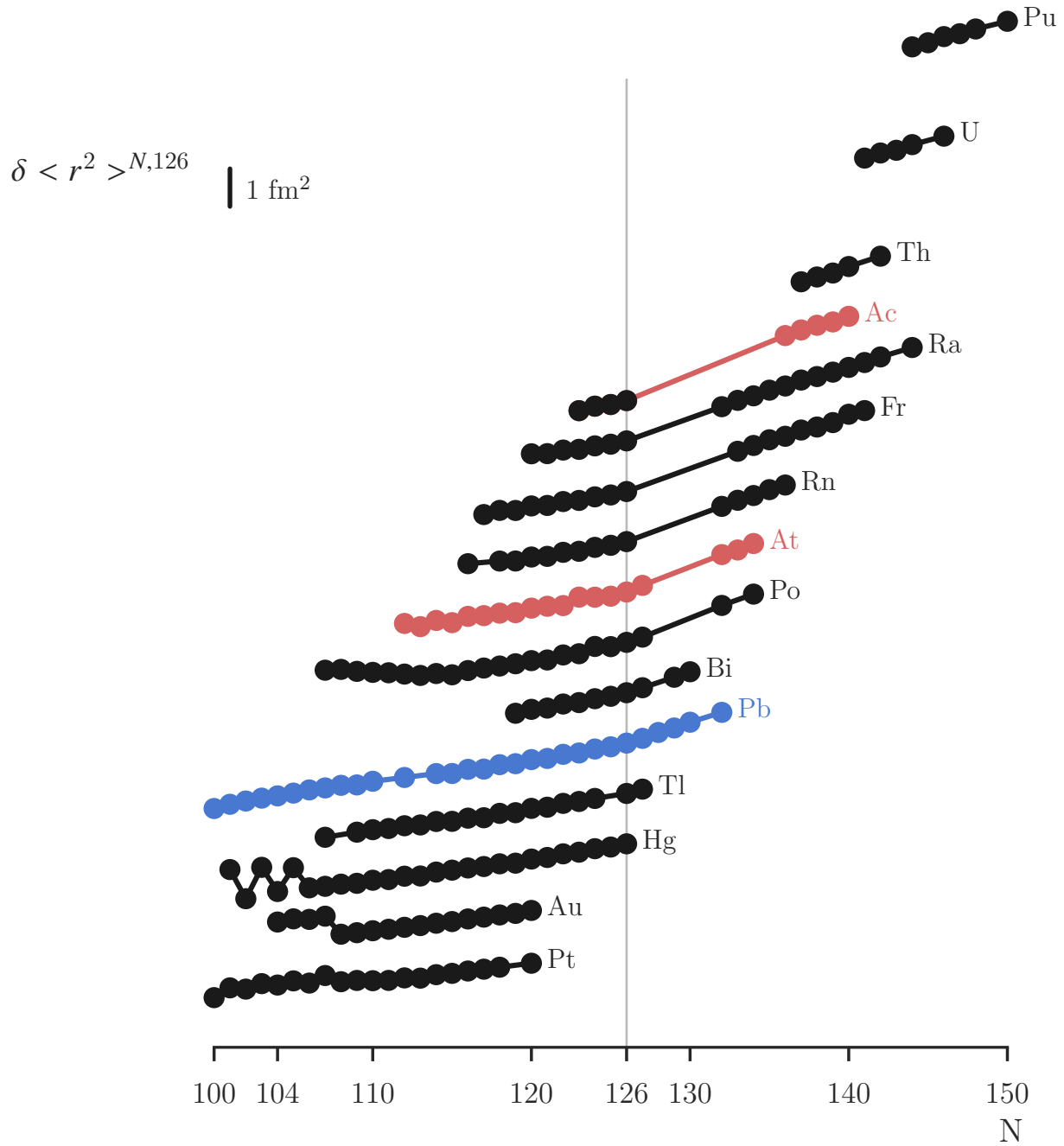


Figure 8.1: Change of the mean squared charge radii $\delta \langle r^2 \rangle$ around the lead isotope chain. $\delta \langle r^2 \rangle$ were detected for the isotopes highlighted in red as a part of in this work. All datasets are separated by 1 fm^2 . The remaining data were extracted from [11, 90].

A | Silver spectra

The isotopes $^{116-119}\text{Ag}$ were analyzed by evaluating their γ spectrum. In order to assign the γ lines in the spectrum to the nuclear ground and isomeric state, or background radiation, all the counts in every observed γ line were investigated as a function of the laser wavelength. All spectra in which a single or double peak resonance can be observed were summarized for the final evaluation. The γ energies used are shown in Figures A.1 to A.4. Not all of those are listed in databases [72, 73].

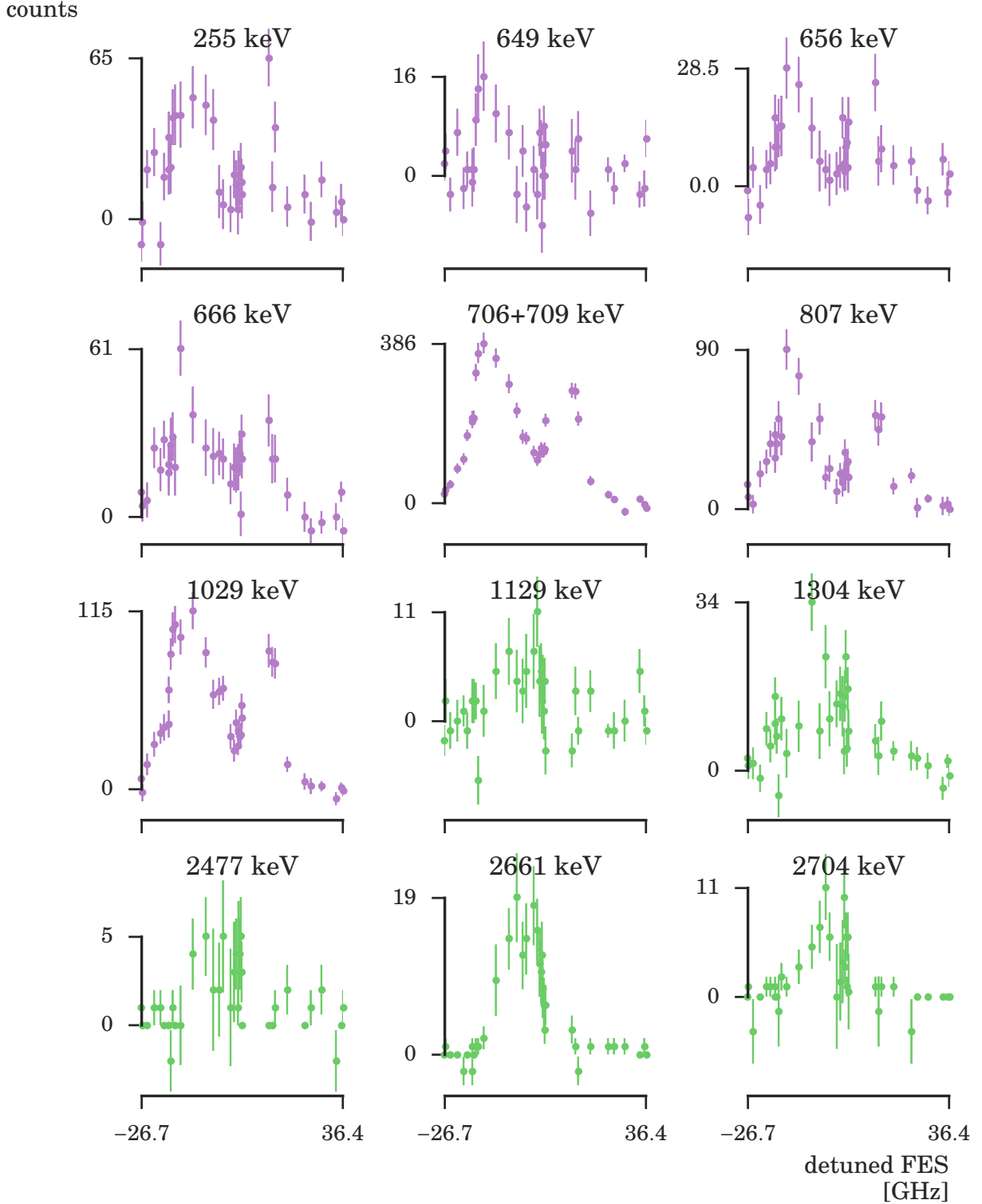


Figure A.1: Raw data for ^{116}Ag evaluation. The data shown in purple was used for the isomer evaluation, and data shown in green for the ground state. Although the γ energies 706 keV and 709 keV can also be found in the ground state, but with branching ratios below 1 % and therefore below the detection limit. 649 keV and 656 keV were not listed in the databases.

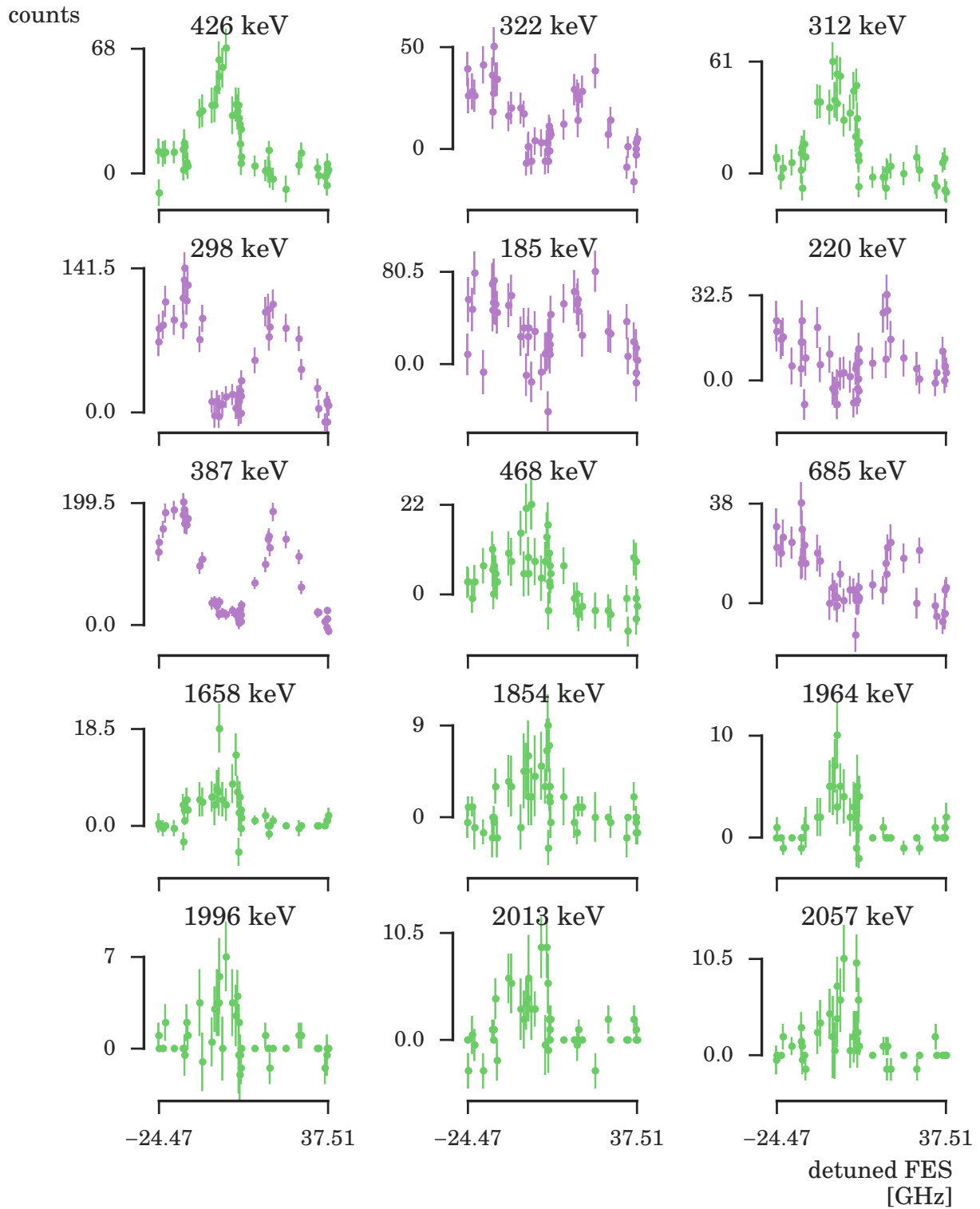


Figure A.2: Raw data for ^{117}Ag evaluation. The data shown in purple was used for the isomer evaluation, and data shown in green for the ground state. The majority of the data marked used for isomer evaluation can also be found in the γ spectrum for the ground state, but with branching ratios below 1%.

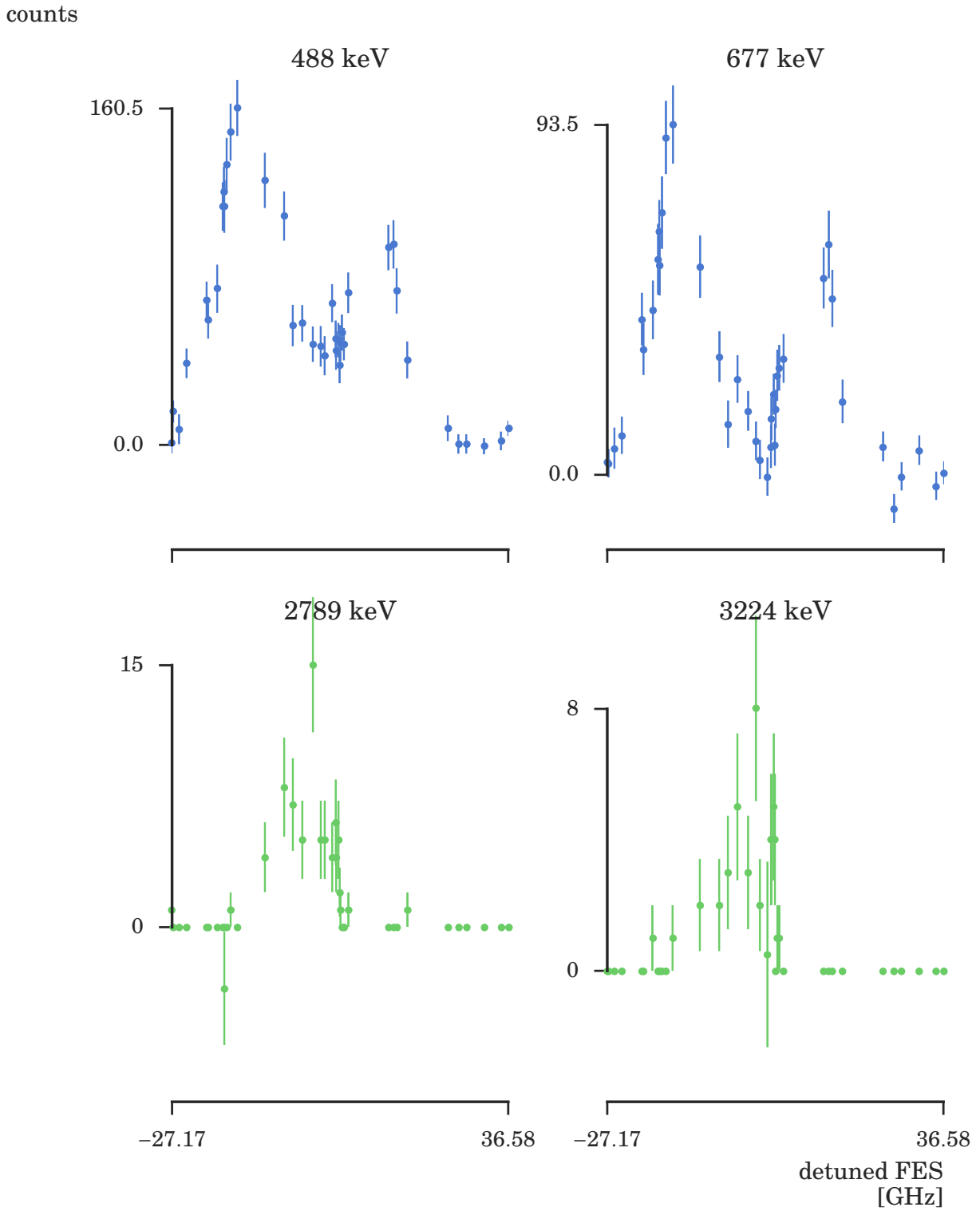


Figure A.3: Raw data for ^{118}Ag evaluation. The data shown in green was used for the ground state evaluation. All γ lines for the isomeric state can also be found in the ground state with similar branching ratios. Those data that can be found in both is indicated in blue.

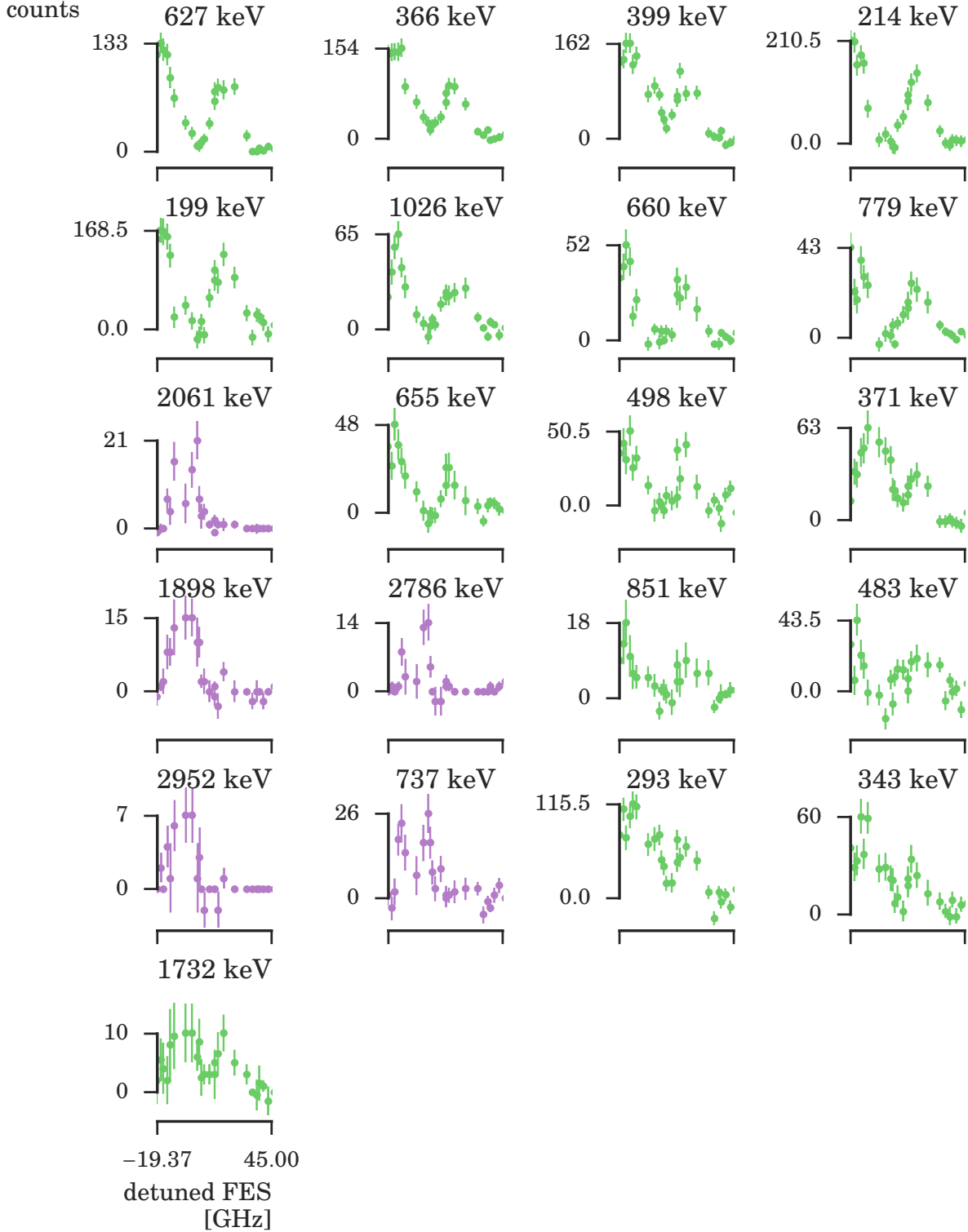


Figure A.4: Raw data for ^{119}Ag evaluation. The data shown in purple was used for the isomer evaluation, and data shown in green for the ground state. In the database [73] no γ energies are listed for the isomeric state are listed. All γ energies marked in purple can be found in the ground state. Although most the data indicated in purple could also show a double-peak spectrum, the dominant peak of these spectra is at the minimum of the green spectrum.

B | Astatine properties

Table B.1: Half-lives, nuclear spin and decay energies for the investigated astatine isotopes. The implantation α -detector is a photo diode that can be moved directly into the RIB, therefore it has a higher detection efficiency, but can only be used for RIBs with short half-lives and low isobaric backgrounds. For ^{199}At an α energy was discovered that shows a laser wavelength dependence. So far it was not possible to clarify whether this α -energy belongs to the nuclear ground state, or an so for undiscovered isomeric state.

isotope	half-life	spin	detector	decay energy [keV]
197g	0.35 s	(9/2)	α implantation	6958
197m	3.7 s	(1/2)	α implantation	6707
198g	4.2 s	(3)	α implantation	6754
198m	1.0 s	(10)	α implantation	6856
199g	4.2 s	(9/2)	α	6643
199m?	?	?	α	6466
200g	43 s	(3)	α	6464
			γ	565
200m1	47 s	(7)	α	6411
			γ	485
200m2	3.5 s	(10)	α	6538
			γ	231 (IT)
201g	89 s	(9/2)	CEM	
202g	184 s	(2,3)	α	6228
202m1	182 s	(7)	α	6135
202m2	0.46 s	(10)	γ	352 (IT)
203g	7.4 min	9/2	α	6087
204g	9.2 min	7	γ	684, 516, 426, 609, 841, 588, 761, 335
			α	5951
205g	26.2 min	9/2	CEM	
			γ	719, 669, 628, 520, 311, 873, 154, 660
				618, 1032, 783, 449, 789, 1325, 385, 1308
			α	5902
206g	30.0 min	(5)	CEM	
207g	1.8 h	9/2	α	5758
			γ	814
208g	1.63 h	6	β	
			γ	687, 660, 178
209g	5.41 h	9/2	α	5647
			γ	545, 552, 786, 792, 195
210g	8.1 h	(5)	β	
			γ	1181, 245, 1483, 1437, 1600
211g	7.21 h	9/2	α	5869
			β	
212g	0.314 s	(1)	α	7616, 7616
212m	0.119 s	(9)	α	7837, 7900
217g	32.3 ms	9/2	α implantation	7066
218g	1.5 s	?	α	6693
219g	56 s	?	α	6275

Acronyms

AI autoionizing state

BBO β -barium-borate

BiBO bismuth-borate

BRF birefringent filter

CEM channeltron electron multiplier

CERN Organisation Européenne pour la Recherche Nucléaire (renamed from Conseil Européen pour la Recherche Nucléaire) (Switzerland)

CoG centre of gravity

cps counts per second

EDM electric dipole moment

FC Faraday cup

FEBIAD forced electron beam induced arc discharge ion source

FES first excitation step

FSR free spectral range

FWHM full-width half-maximum

GANIL Le Grand Accélérateur National d'Ions Lourds (France)

GSI Helmholtzzentrum für Schwerionenforschung (Germany)

HFS hyperfine structure

HPGe high-purity germanium detector

IG-LIS ion guide laser ion source

IP ionization potential

IPN Institut de Physique Nucléaire Orsay (France)

IS isotope shift

ISAC Isotope Separator and ACcelerator

ISOL Isotope Separator On-Line

ISOLDE Isotope Separator On-line DEvice

IT isomeric transition

KDP potassium dihydrogen phosphate

LBO lithum-triborate

LIS laser ion source

LISOL Leuven Isotope Separator On-Line

Nd:YAG neodymium-doped yttrium aluminum garnet

Nd:YVO₄ neodymium-doped yttrium orthovanadate

NIR near infrared

NIST National Institution of Standards and Technology

NMS normal mass shift

NSCL National Superconducting Cyclotron Laboratory (USA)

RADRIIS radioactive decay detection combined with resonance ionization spectroscopy

RFQ radio frequency quadrupole

RIB radioactive ion beam

RIKEN Rikagaku Kenkyujo, The Institute of Physical and Chemical Research (Japan)

SES second excitation step

SIS surface ion source

SMS specific mass shift

SPECT single photon emission computed tomography

TAT targeted alpha therapy

TEM transverse electromagnetic mode

Ti:Sa titanium-doped sapphire

TRILIS TRIUMF's resonant ionization laser ion source

TRIUMF Canada's National Laboratory for Particle and Nuclear Physics and Accelerator-based Science (renamed from Tri University Meson Facility) (Canada)

UV ultraviolet

VIS visible spectrum

Bibliography

- [1] J. J. Thomson. Recombination of gaseous ions, the chemical combination of gases, and monomolecular reactions. *Philos. Mag.*, 47(278):337–378, 1924.
- [2] E. Rutherford. The scattering of α and β particles by matter and the structure of the atom. *Philos. Mag.*, 92(4):379–398, 2012.
- [3] T. Kubo. Recent progress of in-flight separators and rare isotope beam production. *Nucl. Instrum. Methods Phys. Res., Sect. B*, 376:102–110, 2016.
- [4] P. Van Duppen. Isotope Separation On Line and Post Acceleration. In Jim Al-Khalili and Ernst Roeckl, editors, *The Euroschool Lectures on Physics with Exotic Beams, Vol. II*, volume 700, pages 37–77. Springer Berlin Heidelberg, 2006.
- [5] H.J. Kluge, F. Ames, W. Ruster, and K. Wallmeroth. Laser ion sources. In *Proceedings of the Accelerated Radioactive Beams workshop, Parksville, Canada*, 1985.
- [6] G. D. Alkhazov, L. K. Batist, A. A. Bykov, V. D. Vitman, V. S. Letokhov, V. I. Mishin, V. N. Panteleyev, S. K. Sekatsky, and V. N. Fedoseyev. Application of a high efficiency selective laser ion source at the IRIS facility. *Nucl. Instrum. Methods Phys. Res., Sect. A*, 306(1):400 – 402, 1991.
- [7] B. A. Marsh. Resonance ionization laser ion sources for on-line isotope separators. *Rev. Sci. Instrum.*, 85(2):02B923, 2014.

- [8] A. Kramida, Yu. Ralchenko, J. Reader, and NIST ASD Team. NIST Atomic Spectra Database (ver. 5.3), [Online]. <http://physics.nist.gov/asd> [2015, December 15]. National Institute of Standards and Technology, Gaithersburg, MD, 2015.
- [9] P. L. Smith, C. Heise, J. R. Esmond, and R. L. Kurucz. Atomic spectral line database from CD-ROM 23 of R. L. Kurucz. <http://www.pmp.uni-hannover.de/cgi-bin/ssi/test/kurucz/sekur.html>.
- [10] J. Billowes and P. Campbell. High-resolution laser spectroscopy for the study of nuclear sizes and shapes. *J. Phys. G: Nucl. Part. Phys.*, 21(6):707–739, 1995.
- [11] I. Angeli and K. P. Marinova. Table of experimental nuclear ground state charge radii: An update. *At. Data Nucl. Data Tables*, 99(1):69–95, 2013.
- [12] H. H. Stroke, D. Proetel, and H. J. Kluge. Odd-even staggering in mercury isotope shifts: Evidence for Coriolis effects in particle-core coupling. *Phys. Lett. B*, 82(2):204–207, 1979.
- [13] T. E. Cocolios, W. Dexters, M. D. Seliverstov, A. N. Andreyev, S. Antalic, A. E. Barzakh, B. Bastin, J. Büscher, I. G. Darby, D. Fedorov, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, S. Fritzsche, G. Huber, M. Huyse, M. Keupers, U. Köster, Yu. Kudryavtsev, E. Mané, B. A. Marsh, P. L. Molkanov, R. D. Page, A. M. Sjoedin, I. Stefan, J. Van de Walle, P. Van Duppen, M. Venhart, S. G. Zemlyanoy, M. Bender, and P.-H. Heenen. Early Onset of Ground State Deformation in Neutron Deficient Polonium Isotopes. *Phys. Rev. Lett.*, 106(5):052503, 2011.
- [14] <http://www.triumf.ca/research-program/research-facilities/isac-facilities>.
- [15] F. Ames, P. Bricault, H. Heggen, P. Kunz, J. Lassen, A. Mjos, S. Raeder, and A. Teigelhöfer. Ion source developments for the production of radioactive isotope beams at TRIUMF. *Rev. Sci. Instrum.*, 85(2):02B912, 2014.
- [16] A. Osa, M. Matsuda, T. Sato, and S. Ichikawa. Release and efficiencies of radioactive ion beams from the JAEA-ISOL target/ion source system. *Nucl. Instr. and Meth. in Phys. Res. B*, 266(19):4373–4377, 2008.

- [17] U. Köster. *Ausbeute und Spektroskopie radioaktiver Isotope bei LOHENGRIN und ISOLDE*. PhD thesis, Technische Universität München, 2000.
- [18] A. Gallant, J. C. Bale, T. Brunner, U. Chowshury, S. Ettenauer, A. Lennarz, D. Robertson, V. V. Simon, A. Chaudhuri, J. D. Holt, A. A. Kwiatkowski, E. Mané, J. Menéndes, B. E. Schultz, M. C. Simon, C. Andreoiu, P. Delheij, M. R. Pearson, H. Savajols, A. Schwenk, and J. Dilling. New precision mass measurements of neutron-rich calcium and potassium isotopes and three-nucleon forces. *Phys. Rev. Lett.*, 109(3):032506, 2012.
- [19] S. Raeder, M. Dombsky, H. Heggen, J. Lassen, Th. Quenzel, A. M. Sjödin, A. Teigelhöfer, and K. Wendt. In-Source laser spectroscopy developments at TRILIS – Towards spectroscopy on actinium and scandium. *Hyperfine Interact.*, 216(1-3):33–39, 2013.
- [20] F. Garcia. Calculation of in-target rates for radioactive isotope beam production at TRIUMF. Master’s thesis, Simon Fraser University, 2016.
- [21] I. Langmuir and K. H. Kingdon. Thermionic Effects Caused by Vapours of Alkali Metals. *Proc. R. Soc. Lond. A*, 107(741):61–79, 1925.
- [22] R. Kirchner. Ion sources for radioactive beams and related problems. *Rev. Sci. Instrum.*, 67(3):928–933, 1996.
- [23] K. D. A. Wendt, K. Blaum, C. Geppert, P. Müller, W. Nörtershäuser, A. Schmitt, P. Schumann, N. Trautmann, and B. A. Bushaw. Laser based techniques for ultra trace isotope production, spectroscopy and detection. *Hyperfine Interact.*, 162(1):147–157, 2005.
- [24] J. Meißner. TRILIS laser beam transport. Master’s thesis, Hochschule Emden Leer, 2013.
- [25] S. Raeder, H. Heggen, J. Lassen, F. Ames, D. Bishop, P. Bricault, P. Kunz, A. Mjos, and A. Teigelhöfer. An ion guide laser ion source for isobar-suppressed rare isotope beams. *Rev. Sci. Instrum.*, 85(3):033309, 2014.

- [26] A. Sen, F. Ames, P. Bricault, J. Lassen, A. Laxdal, and A. Mjos. Extraction and low energy beam transport from a surface ion source at the TRIUMF-ISAC facility. *Nucl. Instrum. Methods Phys. Res., Sect. B*, 376:97–101, 2016.
- [27] P. G. Bricault, F. Ames, M. Dombsky, P. Kunz, and J. Lassen. Rare isotope beams at ISAC - target & ion source systems. *Hyperfine Interact.*, 225(4):25–49, 2013.
- [28] F. Ames. private communication, 2016.
- [29] P. Kunz, C. Andreoiu, P. Bricault, M. Dombsky, J. Lassen, A. Teigelhöfer, H. Heggen, and F. Wong. Nuclear and in-source laser spectroscopy with the ISAC yield station. *Rev. Sci. Instrum.*, 85(5):053305, 2014.
- [30] G. F. Knoll. *Radiation Detection and Measurement*. Wiley, 4th edition, 2000.
- [31] W. Demtröder. *Laser Spectroscopy: Vol. 2: Experimental Techniques*. Springer, 4th edition, 2008.
- [32] R. V. Ambartzumian and V. S. Letokhov. Selective two-step (STS) photoionization of atoms and photodissociation of molecules by laser radiation. *Appl. Opt.*, 11(2):354–358, 1972.
- [33] W. Demtröder. *Atoms, Molecules and Photons: An Introduction to Atomic -, Molecular- and Quantum Physics*. Springer, Berlin, 1st edition, 2006.
- [34] I. V. Hertel and C.-P. Schulz. *Atome, Moleküle und optische Physik 1: Atomphysik und Grundlagen der Spektroskopie*. Springer, Berlin, 1st edition, 2007. (German).
- [35] R. Li, J. Lassen, A. Teigelhöfer, J. P. Lavoie, P. Bricault, O. Chachkova, J. Meissner, and Zlateva. Resonant Ionization Laser Ion source (RILIS) off-line developments on Ga, Al and Ca. *Nucl. Instrum. Methods Phys. Res., Sect. B*, 308:74–79, 2013.
- [36] U. Fano. Effects of Configuration Interaction on Intensities and Phase Shifts. *Phys. Rev.*, 124(6):1866, 1961.
- [37] J.-P. Connerade. *Highly Excited Atoms*. Cambridge University Press, 1998.

- [38] B. Bushaw, W. Nörtershäuser, K. Blaum, and K. Wendt. Studies of narrow autoionizing resonances in gadolinium. *Spectrochim. Acta, Part B*, 58(6):1083–1095, 2003.
- [39] J. Levine. A simplified calculation of power-broadened linewidth, with application to resonance ionization mass spectrometry. *Spectrochim. Acta, Part B*, 69:61–66, 2012.
- [40] W. Demtröder. *Laser Spectroscopy: Vol. 1: Basic Principles*. Springer, 4th edition, 2008.
- [41] B. Bushaw, W. Nörtershäuser, and K. Wendt. Lineshapes and optical selectivity in high-resolution double-resonance ionization mass spectrometry. *Spectrochim. Acta, Part B*, 54(2):321–332, 1999.
- [42] W. Nörtershäuser, B. Bushaw, P. Müller, and K. Wendt. Line shapes in triple-resonance ionization spectroscopy. *Appl. Opt.*, 39(30):5590–5600, 2000.
- [43] M. Goeppert Mayer. Nuclear Configurations in the Spin-Orbit Coupling Model. I. Empirical Evidence. *Phys. Rev.*, 78(1):16–21, 1950.
- [44] I. I. Sobelman. *Atomic Spectra and Radiative Transitions*. Springer, 1991.
- [45] H. Haken, H. Ch. Wolf, and W. D. Brewer. *The Physics of Atoms and Quanta: Introduction to Experiments and Theory*. Springer-Verlag, 7th edition, 2005.
- [46] P. Stransky, A. Frank, and R. Bijker. On prolate shape predominance in nuclear deformation. *J. Phys. Conf. Ser.*, 322(1):012018, 2011.
- [47] R. Neugart and G. Neyens. Nuclear moments. In Jim Al-Khalili and Ernst Roeckl, editors, *The Euroschool Lectures on Physics with Exotic Beams, Vol. II*, volume 700, pages 135–189. Springer Berlin Heidelberg, 2006.
- [48] R. F. Casten. *Nuclear Structure From A Simple Perspective*. Oxford University Press, 1990.
- [49] W. H. King. *Isotope Shifts in Atomic Spectra*. Springer, 1st edition, 1984.

- [50] W. Nörtershäuser and C. Geppert. Nuclear Charge Radii of Light Elements and Recent Development in Collinear Laser Spectroscopy. In C. Scheidenberger and Marek Pfützner, editors, *The Euroschool Lectures on Physics with Exotic Beams, Vol. IV*, volume 879. Springer Berlin Heidelberg, 2014.
- [51] B Cheal, T. Cocolios, and S. Fritzsche. Laser spectroscopy of radioactive isotopes: Role and limitations of accurate isotope-shift calculations. *Phys. Rev. A*, 86(4):042501, 2012.
- [52] G. W. F. Drake, W. Nörtershäuser, and Z. C. Yan. Isotope shifts and nuclear radius measurements for helium and lithium. *Can. J. Phys.*, 83(4):311–325, 2005.
- [53] A. D. Bromley, editor. *Treatise on Heavy-Ion Science - Nuclei Far From Stability*, volume 8. Plenum Press, New York, 1989.
- [54] E.C. Seltzer. K X-ray Isotope Shifts. *Phys. Rev.*, 188(4):1916, 1969.
- [55] U. Köster, V. N. Fedoseyev, and V. I. Mishin. Resonant laser ionization of radioactive atoms. *Spectrochim. Acta, Part B*, 58(6):1047–1068, 2003.
- [56] V. N. Fedosseev, L. E. Berg, D. V. Fedorov, D. A. Fink, O. J. Launila, R. Losito, B. A. Marsh, R. E. Rossel, S. Rothe, M. D. Seliverstov, A. M. Sjödin, and K. D. A. Wendt. Upgrade of the resonance ionization laser ion source at ISOLDE on-line isotope separation facility: New lasers and new ion beams. *Rev. Sci. Instrum.*, 83(2):02A903, 2012.
- [57] I. D. Moore, A. Nieminen, J. Billowes, P. Campbell, C. Geppert, A. Jokinen, T. Kessler, B. A. Marsh, H. Penttilä, S. Rinta-Antila, B. Tordoff, K. Wendt, and Äystö J. Development of a laser ion source at IGISOL. *J. Phys. G: Nucl. Part. Phys.*, 31(10):1499–1502, 2005.
- [58] V. Sonnenschein, I. D. Moore, I. Pohjalainen, M. Reponen, S. Rothe, and K. Wendt. Intracavity Frequency Doubling and Difference Frequency Mixing for Pulsed ns Ti:sapphire Laser Systems at On-Line Radioactive Ion Beam Facilities. *JPS Conf. Proc.*, 6(3):030126, 2015.

- [59] S. Rothe. Aufbau eines Chrom:Forsterit-Lasers und Resonanzionisationsspektroskopie an Strontium, Titan, Nickel, Scandium und Silicium. Master's thesis, Johannes Gutenberg-Universität Mainz, 2009. (German).
- [60] P. F. Moulton. Spectroscopic and laser characteristics of $\text{Ti:Al}_2\text{O}_3$. *J. Opt. Soc. Am. B*, 3(1):125, 1986.
- [61] W. Koechner. *Solid-State Laser Engineering*. Springer, 6th edition, 2006.
- [62] R. E. Horn. *Aufbau eines Systems gepulster, abstimmbarer Festkörperlaser zum Einsatz in der Resonanzionisations-Massenspektrometrie*. PhD thesis, Johannes Gutenberg-Universität Mainz, 2003. (German).
- [63] E. Hecht. *Optics*. Addison Wesley, 4th edition, 2001.
- [64] D. Albers. Design, Assembly and Characterization of a Double-Sided-Pumped, High Repetition Rate Titanium Sapphire Laser System. Master's thesis, Fachhochschule Oldenburg Ostfriesland Wilhelmshaven, 2007.
- [65] B. Struve. *Laser: Grundlagen, Komponenten, Technik*. Verlag Techn., 2001. (German).
- [66] Spectra-Physics. *Navigator II - Fiber-Coupled, Diode- Pumped, Q -Switched Laser Series YHP70 and YHP40*, 2002. Manual.
- [67] W. Lauth, H. Backe, M. Dahlinger, I. Kluft, P. Schwamb, G. Schwickert, N. Trautmann, and U. Othmer. Resonance ionization spectroscopy in a buffer gas cell with radioactive decay detection, demonstrated using ^{208}Tl . *Phys. Rev. Lett.*, 68(11):1675–1678, 1992.
- [68] R. Ferrer, N. Bree, T. E. Cocolios, I. G. Darby, H. De Witte, W. Dexters, J. Diriken, J. El-seviers, S. Franchoo, M. Huyse, N. Kesteloot, Y. Kudryavtsev, D. Pauwels, D. Radulov, T. Roger, H. Savajols, P. Van Duppen, and M. Venhart. In-gas-cell laser ionization spectroscopy in the vicinity of ^{100}Sn : Magnetic moments and mean-square charge radii of $N=50-54$ Ag. *Phys. Lett. B*, 728:191–197, 2014.

- [69] U. Dinger, J. Eberz, G. Huber, R. Menges, R. Kirchner, O. Klepper, T. Köhl, and D. Marx. Nuclear moments and charge in the charge radii of neutron-deficient silver isotopes. *Nucl. Phys. A*, 503(2):331–348, 1989.
- [70] V. Sebastian. *Laserionisation und Laserionenquelle an ISOLDE/CERN*. PhD thesis, Johannes Gutenberg-Universität Mainz, 1999. (German).
- [71] E. B. Saloman. A resonance ionization spectroscopy resonance ionization mass-spectrometry data service .4. Data sheets for Be, In, Li, K, Rb, Ag, Tl and V and an update of the data sheet for Ni. *Spectrochim. Acta, Part B*, 48(9):1139, 1993.
- [72] R. R. Kinsey et al. The NUDAT/PCNUDAT Program for Nuclear Data, paper submitted to the 9th International Symposium of Capture Gamma-Ray Spectroscopy and Related Topics, Budapest, Hungary, October 1996. Data extracted from the NUDAT database, version 2.6,.
- [73] S. Y. F. Chu, L. P. Ekström, and R. B. Firestone. The Lund/LBNL Nuclear Data Search Version 2.0. <http://nucleardata.nuclear.lu.se/toi/>, 1999.
- [74] *High Finesse - Wavelength meter Ångström WS/7 MC8 Super precision User Manual*.
- [75] W. R. Leo. *Techniques for Nuclear and Particle Physics Experiments - A How to Approach*. Springer Berlin Heidelberg, 1994.
- [76] N. J. Stone. Table of nuclear magnetic dipole and electric quadrupole moments. *At. Data Nucl. Data Tables*, 90(1):75–176, 2005.
- [77] H. Dahmen and S. Penselin. Measurement of the Nuclear Magnetic Dipole Moment of Au^{197} and Hyperfine Structure Measurement in the Ground States of Au^{197} , Ag^{107} , Ag^{109} , and K^{39} . *Z. Phys.*, 200(4):456–466, 1967.
- [78] Y.-S. Kim and M. W. Brechbiel. An overview of targeted alpha therapy. *Tumor Biol.*, 33(3):573–590, 2012.

- [79] P. E. Borchardt, R. R. Yuan, M. Miederer, M. R. McDevitt, and D. A. Scheinberg. Targeted Actinium-225 in Vivo Generators for Therapy of Ovarian Cancer. *Cancer Res.*, 63(16):5084–5090, 2003.
- [80] H. S. Chan, M. W. Konijnenberg, S. de Blois, E. and Koelewijn, R. P. Baum, A. Morgenstern, F. Bruchertseifer, W. A. Breeman, and M. de Jong. Influence of tumour size on the efficacy of targeted alpha therapy with ^{213}Bi -[DOTA0,Tyr3]-octreotate. *EJNMMI Res.*, 6(1):1, 2016.
- [81] S. Agostinelli et al. Geant4 – a simulation toolkit. *Nucl. Instrum. Methods Phys. Res., Sect. A*, 506(3):250 – 303, 2003.
- [82] W. F. Meggers. The spectrochemistry of actinium. *Spectrochim. Acta*, 10(2):195–200, 1957.
- [83] W. F. Meggers, M. Fred, and F. S. Tomkins. Emission Spectra of Actinium. *J. Res. Nat. Bur. Stand.*, 58(6):207–315, 1957.
- [84] J. Roßnagel. Aufbau einer Atomstrahl-Massenspektrometer-Apparatur zur resonanten Laserionisation. Master’s thesis, Johannes Gutenberg-Universität, 2011. (German).
- [85] J. Roßnagel, S. Raeder, A. Hakimi, R. Ferrer, N. Trautmann, and K. Wendt. Determination of the First Ionization Potential of Actinium. *Phys. Rev. A*, 85(1):012525, 2012.
- [86] S. Raeder, H. Savajols, S. Sels, E. Traykov, P. E. Van Den Bergh, S. Sami, C. Van Beveren, P. Van Duppen, K. Wendt, S. Zadvornaya, R. Ferrer, B. Bastin, P. Creemers, P. Delahaye, X. Fléchar, S. Franchoo, L. Ghys, L. Gaffney, C. Granados, R. Heink, L. Hijazi, M. Huyse, Y. Kudryavtsev, M. Laatiaoui, N. Lecesne, F. Lutton, I. Moore, Y. Martinez, E. Mogilevskiy, P. Naubereit, J. Piot, and S. Rothe. In-gas-jet spectroscopy of actinium isotopes. In *EMIS XVII - International Conference on Electromagnetic Isotope Separators and Related Topics*, Grand Rapids, United States, 2015.
- [87] S. Raeder. private communication, 2014.

- [88] V. Sonnenschein. *Laser Development and High Resolution Resonance Ionization Spectroscopy of Actinide Elements*. PhD thesis, University of Jyväskylä, 2015.
- [89] F. S. Tomkins, M. Fred, and W. F. Meggers. Nuclear Spin of Actinium 227. *Phys. Rev.*, 84(1):168–168, 1951.
- [90] R. Ferrer, C. A. Granados-Buitrago, S. Raeder, et al. Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion. *Nat. Commun.*, 8:14520, 2017.
- [91] S. L. Kaufman. High-resolution laser spectroscopy in fast beams. *Opt. Commun.*, 17(3):309–312, 1976.
- [92] E. R. Tardiff, E. T. Rand, G. C. Ball, T. E. Chupp, A. B. Garnsworthy, P. E. Garrett, M. E. Hayden, C. A. Kierans, W. Lorenzon, M. R. Pearson, C. Schaub, and C. E. Svensson. The radon EDM apparatus. In *ISAC and ARIEL: The TRIUMF Radioactive Beam Facilities and the Scientific Program*, pages 197–206. Springer, 2013.
- [93] B. Garner, Y. Chen, E. G. Lindahl, and B. R. Heckel. Reduced Limit on the Permanent Electric Dipole Moment of ^{199}Hg . *Phys. Rev. Lett.*, 116(16):161601, 2016.
- [94] B. Karlik and T. Bernert. Das Element 85 in den natürlichen Zerfallsreihen. *Z. Phys.*, 123(1-2):51–72, 1944. (German).
- [95] R. McLaughlin. Absorption Spectrum of Astatine. *J. Opt. Soc. Am.*, 54(8):965–967, 1964.
- [96] A. Teigelhöfer. Astatine and Yttrium Resonant Ionization Laser Spectroscopy. Master’s thesis, University of Manitoba, 2012.
- [97] S. Rothe, A. N. Andreyev, S. Antalic, A. Borschevsky, L. Capponi, T. E. Cocolios, H. De Witte, E. Eliav, D. V. Fedorov, V. N. Fedosseev, D. A. Fink, S. Fritzsche, L. Ghys, M. Huyse, N. Imai, U. Kaldor, Y. Kudryavtsev, U. Köster, J. F. W. Lane, J. Lassen, V. Liberati, Lynch K. M., B. A. Marsh, K. Nishio, D. Pauwels, V. Pershina, L. Popescu, T.J. Procter,

- D. Radulov, S. Raeder, M. M. Rajabali, Rapisarda E., R. E. Rossel, K. Sandhu, M. D. Seliverstov, A. M. Sjoedin, P. Van den Bergh, P. Van Duppen, M. Venhart, Y. Wakabayashi, and K. D. A. Wendt. Measurement of the first ionization potential of astatine by laser ionization spectroscopy. *Nat. Commun.*, 4:1835, 2013.
- [98] A. Borschevsky. private communication, 2016.
- [99] P. Campbell, J. Billowes, J. A. Behr, G. Gwinner, G. D. Sprouse, and F. Xu. Isotope shifts of the neutron-deficient bismuth isotopes: charge radii systematics across the $Z=82$ shell closure. *Phys. Lett. B*, 346(1):21–26, 1995.
- [100] R. Menges, U. Dinger, N. Boos, G. Huber, S. Schröder, S. Dutta, R. Kirchner, O. Klepper, T. Kühn, D. Marx, and G. D. Sprouse. Nuclear moments and the change in the mean square charge radius of neutron deficient thallium isotopes. *Z. Phys. A*, 341(4):475–479, 1992.
- [101] R. Neugart, H. H. Stroke, S. A. Ahmad, H. T. Duong, H. L. Ravn, and K. Wendt. Nuclear Magnetic Moment of ^{207}Tl . *Phys. Rev. Lett.*, 55(15):1559–1562, 1985.
- [102] H. A. Schuessler, E. C. Benck, F. Buchinger, and H. K. Carter. Nuclear spectroscopy using lasers at Oak Ridge National Laboratory: experiments with stable (past) and radioactive (future) tandem beams. *Nucl. Instrum. Methods Phys. Res., Sect. A*, 352(3):583 – 587, 1995.
- [103] J. A. Bounds, C. R. Bingham, H. K. Carter, G. A. Leander, R. L. Mlekodaj, E. H. Spejewski, and W. M. Fairbank. Nuclear structure of light thallium isotopes as deduced from laser spectroscopy on a fast atom beam. *Phys. Rev. C*, 36(5):2560–2568, 1987.
- [104] M. Anselment, W. Faubel, S. Göring, A. Hanser, G. Meisel, H. Rebel, and G. Schatz. The odd-even staggering of the nuclear charge radii of Pb isotopes. *Nucl. Phys. A*, 451(3):471 – 480, 1986.
- [105] M. D. Seliverstov, A. N. Andreyev, N. Barré, A. E. Barzakh, S. Dean, H. De Witte, D. V. Fedorov, V. N. Fedoseyev, L. M. Fraile, S. Franchoo, J. Genevey, G. Huber, M. Huyse, U. Köster, P. Kunz, S. R. Leshner, B. A. Marsh, I. Mukha, B. Roussière, J. Sauvage, I. Ste-

- fanescu, K. Van de Vel, P. Van Duppen, and Yu. M. Volkov. Charge radii and magnetic moments of odd- A $^{183-189}\text{Pb}$ isotopes. *Eur. Phys. J. A*, 41(3):315–321, 2009.
- [106] M. R. Pearson, P. Campbell, K. Leerungrat, J. Billowes, I. S. Grant, M. Keim, J. Kilkallon, I. D. Moore, R. Neugart, M. Neuroth, S. Wilbert, and Isolde Collaboration. Nuclear moments and charge radii of bismuth isotopes. *J. Phys. G: Nucl. Part. Phys.*, 26(12):1829–1848, 2000.
- [107] B. A. Marsh. *In-Source Laser Resonance Ionization at ISOL Facilities*. PhD thesis, University of Manchester, 2007.
- [108] D. Kowalewska, K. Bekk, S. Göring, A. Hanser, W. Kälber, G. Meisel, and H. Rebel. Isotope shifts and hyperfine structure in polonium isotopes by atomic-beam laser spectroscopy. *Phys. Rev. A*, 44(4):R1442–R1445, 1991.
- [109] H. De Witte, A. N. Andreyev, N. Barré, M. Bender, T. E. Cocolios, S. Dean, D. Fedorov, V. N. Fedoseyev, L. M. Fraile, S. Franchoo, V. Hellemans, P. H. Heenen, K. Heyde, G. Huber, M. Huyse, H. Jeppessen, U. Köster, P. Kunz, S. R. Leshner, B. A. Marsh, I. Mukha, B. Rousière, J. Sauvage, M. Seliverstov, I. Stefanescu, E. Tengborn, K. Van de Vel, J. Van de Walle, P. Van Duppen, and Yu. Volkov. Nuclear Charge Radii of Neutron-Deficient Lead Isotopes Beyond $N = 104$ Midshell Investigated by In-Source Laser Spectroscopy. *Phys. Rev. Lett.*, 98(11):112502, 2007.
- [110] W. D. Myers and K. H. Schmidt. An update on droplet model charge distributions. *Nucl. Phys. A*, 410(1):61–73, 1983.
- [111] D. Berdichevsky and F. Tondeur. Nuclear Core Densities, Isotope Shifts, and the Parametrization of the Droplet Model. *Z. Phys. A - Atomic Nuclei*, 322(1):141–147, 1985.
- [112] C. A. Granados-Buitrago. *Investigation of the hyperfine structure of heavy elements by the in-gas laser ionization and spectroscopy*. PhD thesis, Katholieke Universiteit Leuven, 2017. in preparation.

- [113] I. Budinčević, J. Billowes, M. L. Bissell, T. E. Cocolios, R. P. De Groote, S. De Schepper, V. N. Fedosseev, K. T. Flanagan, S. Franchoo, R. F. Garcia Ruiz, H. Heylen, Lynch K. M., B. A. Marsh, G. Neyens, T. J. Procter, R. E. Rossel, S. Rothe, I. Strashnov, H. H. Stroke, and K. D. A. Wendt. Laser spectroscopy of francium isotopes at the borders of the region of reflection asymmetry. *Phys. Rev. C*, 90(1):014317, 2014.