# Nuclear charge radius determination of $^{7,10}\mathrm{Be}$ and the one-neutron halo nucleus $^{11}\mathrm{Be}$

DISSERTATION ZUR ERLANGUNG DES GRADES "Doktor der Naturwissenschaften" Im Promotionsfach Chemie

AM FACHBEREICH CHEMIE, PHARMAZIE UND GEOWISSENSCHAFTEN DER JOHANNES GUTENBERG-UNIVERSITÄT MAINZ

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MAINZ, DEN 30. APRIL 2009

Dekan:

- 1. Berichterstatter:
- 2. Berichterstatter:

Tag der mündlichen Prüfung: 05.06.2009

### Zusammenfassung

Die kernmodellunabhängige Bestimmung der Kernladungsradien kurzlebiger Isotope ist nur durch die Messung der Isotopieverschiebung möglich. Für die leichtesten Elemente (Z < 10) werden hierbei allerdings extreme Anforderungen an die Genauigkeit in Experiment und Theorie gestellt, die bisher nur für He und Li erreicht wurden. Die Ladungsradien der leichtesten Elemente sind von großem Interesse, da in dieser Region sogenannte Halokerne existieren, die sich durch eine sehr exotische Kernstruktur auszeichnen: Sie besitzen einen kompakten Rumpfkern und einen weit ausgedehnten Bereich verdünnter Kernmaterie. Beispiele dafür sind <sup>6</sup>He, <sup>8</sup>He, <sup>11</sup>Li und das in dieser Arbeit untersuchte Isotop <sup>11</sup>Be. Darüberhinaus sind die leichten Elemente auch deshalb von Interesse, weil bislang nur bei solchen Systemen mit wenigen Nukleonen *ab-initio* Berechnungen der Kernstruktur möglich sind. Die Ladungsradien sind hierbei wichtige Vergleichspunkte für diese Theorien. Am Institut für Kernchemie der Johannes Gutenberg-Universität Mainz wurden in dieser Arbeit zwei Ansätze verfolgt, mit denen die Vermessung der Isotopieverschiebung in der D1 Linie für die Isotope <sup>7,10,11</sup>Be<sup>+</sup> mit jeweils unterschiedlicher Genauigkeit erreicht wird.

Der erste Ansatz sieht vor Be-Ionen in einer linearen Paul-Falle zu speichern, dort bis zum Doppler-Limit zu kühlen und schließlich zu spektroskopieren. Damit soll eine Genauigkeit von einigen 100 kHz erreicht werden. Im Rahmen dieser Arbeit wurden zwei lineare Paul Fallen für diesen Zweck entwickelt. Desweiteren wurde die Peripherie, die es ermöglicht Be-Ionen mit einer Startenergie von ca. 60 keV effizient abzubremsen und in die lineare Paul Falle zu transferieren, simuliert und aufgebaut. Für eine der entwickelten Fallen konnte die Speicherung von Ionen bereits demonstriert werden. Der direkte optische Nachweis von gespeicherten <sup>9</sup>Be<sup>+</sup> Ionen wurde jedoch durch die Entwicklungsarbeiten des zweiten experimentellen Ansatzes verzögert und konnte daher im Rahmen dieser Arbeit nicht zum Abschluss gebracht werden.

Dieser zweite Ansatz greift auf die Technik der kollinearen Laserspektroskopie zurück, mit der in den letzten 30 Jahren die Isotopieverschiebung für eine Vielzahl schwerer Isotope bestimmt wurde. Bei leichten Isotopen konnte bisher aber nicht die benötigte Genauigkeit erreicht werden, um Informationen über den Kernladungsradius zu extrahieren. Die Kombination der kollinearen Laserspektroskopie mit modernsten Methoden der Frequenzmeterologie in dieser Arbeit, ermöglichte die erstmalige Bestimmung der Kernladungsradien der Isotope <sup>7,10</sup>Be und des Ein-Neutron-Halo Kerns <sup>11</sup>Be am COLLAPS Experiment an ISOLDE/CERN. Im Rahmen dieser Arbeit konnten die Übergangsfrequenzen und die Isotopieverschiebungen der D1 Linie für die oben genannten Isotope mit einer Genauigkeit von besser als 2 MHz bestimmt werden. In Kombination mit neuesten Berechnungen des Masseneffektes ergaben sich die Kernladungsradien der Isotope <sup>7,10,11</sup>Be mit einem relativen Fehler besser als 1 %. Der Kernladungsradius nimmt von <sup>7</sup>Be zu <sup>10</sup>Be hin kontinuierlich ab und steigt bei <sup>11</sup>Be wieder an. Dieses Resultat wird mit den Vorhersagen verschiedener *ab-initio* Kernmodelle verglichen, die den beobachteten Trend sehr gut widergeben und von denen insbesondere die "Greens Function Monte Carlo" und die "Fermionic Molecular Dynamic" Berechnungen eine sehr gute Übereinstimmung zeigen.

### Abstract

The only nuclear model independent method for the determination of nuclear charge radii of short-lived radioactive isotopes is the measurement of the isotope shift. For light elements (Z < 10) extremely high accuracy in experiment and theory is required and was only reached for He and Li so far. The nuclear charge radii of the lightest elements are of great interest because they have isotopes which exhibit so-called halo nuclei. Those nuclei are characterized by a a very exotic nuclear structure: They have a compact core and an area of less dense nuclear matter that extends far from this core. Examples for halo nuclei are <sup>6</sup>He, <sup>8</sup>He, <sup>11</sup>Li and <sup>11</sup>Be that is investigated in this thesis. Furthermore these isotopes are of interest because up to now only for such systems with a few nucleons the nuclear structure can be calculated *ab-initio*. In the Institut für Kernchemie at the Johannes Gutenberg-Universität Mainz two approaches with different accuracy were developed. The goal of these approaches was the measurement of the isotope shifts between <sup>7,10,11</sup>Be<sup>+</sup> and <sup>9</sup>Be<sup>+</sup> in the D1 line.

The first approach is laser spectroscopy on laser cooled  $Be^+$  ions that are trapped in a linear Paul trap. The accessible accuracy should be in the order of some 100 kHz. In this thesis two types of linear Paul traps were developed for this purpose. Moreover, the peripheral experimental setup was simulated and constructed. It allows the efficient deceleration of fast ions with an initial energy of 60 keV down to some eV and an efficient transport into the ion trap. For one of the Paul traps the ion trapping could already be demonstrated, while the optical detection of captured <sup>9</sup>Be<sup>+</sup> ions could not be completed, because the development work was delayed by the second approach.

The second approach uses the technique of collinear laser spectroscopy that was already applied in the last 30 years for measuring isotope shifts of plenty of heavier isotopes. For light elements (Z < 10), it was so far not possible to reach the accuracy that is required to extract information about nuclear charge radii. The combination of collinear laser spectroscopy with the most modern methods of frequency metrology finally permitted the first-time determination of the nuclear charge radii of <sup>7,10</sup>Be and the one neutron halo nucleus <sup>11</sup>Be at the COLLAPS experiment at ISOLDE/ CERN. In the course of the work reported in this thesis it was possible to measure the absolute transition frequencies and the isotope shifts in the D1 line for the Be isotopes mentioned above with an accuracy of better than 2 MHz. Combination with the most recent calculations of the mass effect allowed the extraction of the nuclear charge radii of <sup>7,10,11</sup>Be with an relative accuracy better than 1 %. The nuclear charge radius decreases from <sup>7</sup>Be continuously to <sup>10</sup>Be and increases again for <sup>11</sup>Be. This result is compared with predictions of *ab-initio* nuclear models which reproduce the observed trend. Particularly the "Greens Function Monte Carlo" and the "Fermionic Molecular Dynamic" model show very good agreement.

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# Chapter 1 Introduction

Since Chadwick discovered the neutron in 1932 it is known that the atomic nucleus of an isotope  ${}^{A}_{N}X^{Z}$  consists of Z protons and N neutrons as already proposed by Rutherford in 1920 [SIM95]. The strong interaction, the force which holds these nucleons together, is still an important research topic. Macroscopic models, like the liquid drop model, which assume a constant mass density lead to a rough estimation of the size of the nucleus, which can be given through the equivalent radius  $R_{s}$  [MAY00]:

$$R_s \sim \sqrt[3]{A} \text{ fm}$$

It is known, that nuclei can show slight deviations from this behavior, either caused by shell effects or, especially on heavy isotopes, due to deformations.

In the 1980s Tanihata et al. determined the nuclear matter radii for the full He, Li and Be isotope chain from interaction cross section measurements [TAN85, TAN88]. Isotopes close to the neutron drip line of those elements have tremendously larger matter radii than the other isotopes of the chain as shown in Figure 1.1. First attempts of an explanation were that these light nuclei must be either deformed or exhibit a long tail in the matter distribution [TAN85]. Following experiments particularly the determination of the magnetic moment of <sup>11</sup>Li [ARN87] excluded a strong deformation of the nucleus since the value was very close to the Schmidt value, indicating a nearly spherical shape. In the seminal paper of Hansen and Jonson [HAN87] the authors introduced the name halo-nuclei and pointed out the importance of the very small binding energy of the valence nucleons for the existence of a halo.

Compared to the typical neutron separation energy of approximately 5 MeV in stable isotopes, the one-neutron separation energy of <sup>11</sup>Be of approximately B = 500 keV is extremely small [SUZ95]. According to [HAN87] this weak binding leads to an exponentially decreasing probability distribution of the halo neutron  $e^{-\frac{r}{\kappa}}$  with a decay length of  $\kappa = \frac{\hbar}{2\mu B} \approx 7$  fm. Here,  $\mu$  is the reduced mass of the <sup>10</sup>Be nucleus and the halo neutron. According to the uncertainty principle this very large spatial distribution leads to a small momentum distribution in neutron separation reactions. For <sup>11</sup>Be this momentum distribution is 80 MeV/c which is very small compared to the Fermi momentum of 300 MeV/c found for ordinary fragmentation reactions. In the example of <sup>11</sup>Li the spatial



Figure 1.1: The RMS matter radii for the full He, Li and Be isotope chain are shown. They were extracted by Glauber-type calculations from scattering experiments performed by [TAN85, TAN88].

distribution for the two halo neutrons is so large, that they spend approximately 80% of the time outside the attractive core potential [HAN87] which indicates that the nature of nuclear binding forces is still not completely understood. All these investigations finally led to the picture of halo nuclei; a strongly bound inner core that is surrounded by halo nucleons which can be either protons or neutrons. For the classification of a halo nucleus three characteristics have to be fulfilled:

- 1. The cross section in nuclear reaction experiments is escalating for one isotope.
- 2. Halo nucleons are loosely bound to the core and have a binding energy that is small compared to the general value of 5 MeV/ nucleon, typically in the order of 1 MeV.
- 3. The products of neutron removal reactions in halo nuclei have narrow momentum distributions, which means extended wave functions due to Heisenberg's uncertainty principle.

Beside the very well examined isotopes of He, Li and Be there are other light elements,



Figure 1.2: Halo nuclei and halo candidates in the lower region of the nuclear chart. Halo nuclei are indicated by the expected number of halo nucleons in the squares. Those short-lived isotopes for which charge radii have been measured previously with laser spectroscopy are indicated by blue and red borders and those tackled in this work have green borders.

shown in figure 1.2, for which these characteristics seem to be fulfilled or are already approved.

For a better understanding of the binding mechanisms between inner core and surrounding nucleons it is essential to investigate in which way the inner core is influenced by the halo. If it is modified in some way, it is obvious that the charge respectively the proton distribution of the core will change. The most common and intuitive way of investigating the nuclear charge distribution is electron scattering. Unfortunately this method needs a target of the isotope of interest which means that short-lived isotopes can not be investigated.

The only model-independent way to determine the nuclear charge radius of short-lived isotopes is the measurement of the isotope shift [OTT89], which has been successfully applied for many nuclei with  $Z \ge 10$  [KLU03]. Approaching lighter elements is a big challenge, since the field shift which contains the information about the charge contribution is 10.000 times smaller than the mass shift between two isotopes. Hence, separation between mass shift and field shift - which is a crucial point already for medium heavy nuclei - becomes extremely important and even small errors in the estimated mass shift will lead to large relative deviations in the field shift. This problem could only be solved when mass shift calculations with spectroscopic accuracy, i.e., relative accuracies in the order a few times  $10^{-6}$  became possible. This accuracy was reached by [DRA95, YAN00] and it became possible to extract the field shift contribution reliably as the difference between calculated mass shift and measured isotope shift. This was the basis for a number of experiments on the isotope chains of helium and lithium.

In the last years it was possible to determine the field shift contribution for the whole He and Li isotope chain. <sup>6,8</sup>He have been measured via spectroscopy of single atoms trapped in a magneto optical trap (MOT) [WAN04, MUE07] and <sup>7,8,9,11</sup>Li have been measured via Doppler-free two-photon spectroscopy [EWA04, SAN06a]. As He and Li have been measured with unmatched accuracy, Be is the next heavier element of interest – particularly the one neutron halo nucleus <sup>11</sup>Be. <sup>11</sup>Be is the prototype of a one neutron halo. The determination of its nuclear charge radius, respectively the interaction between halo and core, will contribute considerably to the understanding of nuclear binding forces and nuclear structure, because an interaction between several halo nucleons cannot occur, which simplifies the theoretical interpretation of the result.

The nuclear charge radius for <sup>7</sup>Be is also of enormous interest in astrophysical studies. Fusion reactions described in the solar standard model and neutrino properties can be probed by measuring the solar neutrino flux. The solar fusion chain has three main components [ARP08], which result in the emission of mono energetic electron neutrinos that can be measured on earth. For a better understanding of fusion reactions in the sun as well as of the neutrino oscillation process, precise calculations of the branching ratio between these three possibilities are needed. The reaction probabilities, respectively the cross sections, for a reaction are also determined by the deformations and the nuclear charge radii of the participating nuclei. A measurement of the nuclear charge radius of <sup>7</sup>Be will provide the standard solar model with new input parameters.

In this work, two different approaches which provide different accuracies for the measurement of the isotope shift of Be isotopes are presented. The first approach led to the acronym for these two experiments: BeTINa (**Be**ryllium **T**rap for the Investigation of **N**uclear charge radii). Here, the idea is to capture Be<sup>+</sup> ions, delivered from a radioactive ion beam source in a linear Paul trap, cool them down to a temperature of some mK by Doppler cooling and to perform precision spectroscopy on these ions. This is a non-trivial task which is also followed by a collaboration at RIKEN. Their original goal was a measurement of the hyperfine anomaly of <sup>7,9,10,11</sup>Be by rf-laser double resonance spectroscopy for more than 10 years now. [OKA98]. This group recently reported improved measurements of the nuclear magnetic moment [OKA08] of <sup>7</sup>Be. After the first measurements of isotope shifts on helium and lithium, they have directed their research also towards a possible measurement of beryllium isotope shifts and reported on first results with radioactive Be isotopes [NAK06] shortly after the BeTINa proposal [ZAK06]. However, the achieved accuracy was still about one to two orders of magnitude worse than required for the determination of the nuclear charge radii.

At about the same time, first results of Be<sup>+</sup> isotope shift calculations were obtained. The size of the field shift coefficient that determines the sensitivity of a transition to changes of the charge radius indicated that an accuracy in the order of 1 MHz should be sufficient for a determination of the charge radius on a level of 1%. These calculations triggered a new approach for BeTINa. It was concluded that collinear laser spectroscopy could offer the required accuracy if the limitations due to inaccurate knowledge of the acceleration voltage are eliminated. For this purpose a measurement of the absolute transition frequencies in collinear and anti-collinear geometry was proposed [NOE07]. Thus, the feasibility of this concept had to be demonstrated first in off-line measurements with the stable isotope <sup>9</sup>Be. Parallel to these developments and tests the original approach of trap measurements was continued just in case that the collinear technique would not work sufficiently well. Additionally, the trapped ions would allow to perform more accurate isotope shift or hyperfine anomaly measurements later on.

For these reasons this work is split into two parts:

- the design, construction and first tests of an rf trap system for the spectroscopy of Be<sup>+</sup> ions including the ion optics for deceleration and capturing of ions produced at the ISOLDE facility.
- tests of the collinear/ anti-collinear approach at the existing COLLAPS beam line at ISOLDE, which was adopted to the needs of our experiment. It lead to the first measurement of the nuclear charge radii of <sup>7,10,11</sup>Be.

The thesis is structured in the following way: It starts with some background information about the material beryllium. Chapter 3 explains basic ideas in atomic physics, which are required for both experimental approaches. Chapter 4 and 5 describe trapping and cooling of ions as well as spectroscopy on fast ions and clarify limitations in both cases and how these limitations can be overcome. In chapter 6 the experimental setup for the determination of the nuclear charge radii at the COLLAPS beam line is explained in more detail and the achieved results are presented. In chapter 7 the approach of high resolution spectroscopy on laser cooled Be<sup>+</sup> ions at ISOLTRAP is summarized, while chapter 8 describes the developed Paul traps and first experimental results for trapping of Li<sup>+</sup> and residual gas ions. The work finishes with a summary of both experiments and future perspectives.

CHAPTER 1. INTRODUCTION

## Chapter 2

# Bulk and atomic properties of beryllium

### 2.1 Appearance, production and usage

Beryllium is a silver gray earth alkali metal. It was discovered in 1798 from Louis Nicolas Vaaqueli who separated beryllium oxide from beryl and the gemstone emerald. Not till hundred years later Paul Marie Alfred Lebeau was able to isolate pure beryllium metal through hot electrolysis (Schmelzflusselektrolyse) of  $Na_2[BeF_2]^1$ .

In former times the gemstone beryl was used to manufacture simple lenses and later on glasses which finally led to the German word for glasses: "Brille". Together with an element that emits  $\alpha$ -particles beryllium acts as a neutron emitter

$${}^{9}_{4}\text{Be} + {}^{4}_{2}\text{He} \rightarrow {}^{12}_{6}\text{C} + {}^{1}_{0}\text{n}$$
 (2.1)

and was used together with polonium as ignition for the nuclear bomb released above Hiroshima in 1945. Nowadays beryllium is used in many different areas in science and industry because it is a light metal that combines a high melting point, high heat capacity, high elasticity modulus and a high damping factor. Some physical properties that are needed in this thesis are listed in table 2.1. The mass value for <sup>11</sup>Be is taken from [RIN09], which presents the most accurate mass measurements of Be isotopes and was cited in advance by [PUC09]. Also the masses for <sup>9,10</sup>Be were measured with higher accuracy and deviations of about  $2\sigma$  were found in comparison to the AME mass evaluation, but for data analysis the AME values were used, since [RIN09] was not published.

#### 2.2 Safety concern

Beryllium, beryllium oxides and its salts are extremely toxic and carcinogenic. If inappropriately used, lung, liver and milt damages can occur. To avoid allergenic reactions, skin contact with beryllium should be avoided. The inhalation of beryllium and all its compounds is most dangerous and has to be avoided by all means. In Germany the maximum

 $<sup>^{1}</sup>$ Natriumfluoroberyllat

Isotope	Ι	$\mathbf{T}_{1/2}$	Decay	Mass	Source
$^{7}$ Be	3/2	53 d	$\beta^+$	7.01692983(11)	[AUD03]
<sup>9</sup> Be	3/2	stable		9.01218220(43)	[AUD03]
$^{10}$ Be	0	$1.5 \cdot 10^6$ years	$\beta^{-}$	10.01353382(43)	[AUD03]
$^{11}$ Be	1/2	13.8 sec	$\beta^{-}$	11.02166155(62)	[RIN09]
$^{12}$ Be	0	$23.6 \mathrm{msec}$	$\beta^{-}$	12.026921(16)	[AUD03]
$^{14}$ Be	0	$4.3 \mathrm{msec}$	$\beta^{-}$	14.042890(140)	[AUD03]

Table 2.1: Atomic properties of beryllium isotopes. Nuclear spin I, half-life  $t_{1/2}$ , decay mode and atomic masses are listed.

concentration of Be around the working place is specified in the so called MAK value (MAK - Maximale Arbeitsplatzkonzentration) and is 0.002  $\frac{\text{mg}}{\text{m}^3}$ . That means the working area for an eight hour working day five days a week has to contain less than  $2.2 \cdot 10^{-7} \frac{\text{mol}}{\text{m}^3}$  of Be. But as mentioned in [KAM08] also a short exposure to peak concentrations can dramatically increase the risk of severe health consequences.

# Chapter 3

## Atomic structure theory

The interaction of nuclear electromagnetic moments with the electron shell results in more or less complicated spectra, exhibiting resolved or unresolved hyperfine structure for those isotopes with angular spin  $I \neq 0$  which can nowadays be resolved thanks to the development of narrow linewidth laser sources. For a better understanding of the spectra obtained during the measurements at ISOLDE and the precautions which have to be considered for laser cooling and spectroscopy of <sup>9</sup>Be<sup>+</sup> in a linear Paul trap, the following section describes the hyperfine splitting of beryllium isotopes and the resulting consequences based on [MAY00, DRA96].

#### **3.1** Hyperfine structure

A nucleus with a non-vanishing spin exhibits a magnetic moment and those which are additionally deformed also an electric quadrupole moment. Both interact with the magnetic and electric fields of the electron shell at the nuclear site. This leads to a coupling of the nuclear spin I with the angular momentum J of the electron shell to the total angular momentum F and to a splitting of the atomic energy levels, which is called hyperfine splitting  $\Delta E_{HFS}$ .  $\Delta E_{HFS}$  is the sum of the interaction energy  $\Delta E_M$  between the magnetic moments of nucleus and shell

$$\Delta E_M = \frac{A}{2}C\tag{3.1}$$

and of the interaction energy  $\Delta E_Q$  between the electric quadrupole moment of the core with the electric field gradient caused by electrons at the nucleus

$$\Delta E_Q = \frac{B}{4} \left[ \frac{3}{2} \frac{C(C+1) - 2I(I+1) \cdot J(J+1)}{(2I-1) \cdot (2J-1) \cdot I \cdot J} \right], \tag{3.2}$$

with C = F(F+1) - I(I+1) - J(J+1). The coupling constant A is also called *interval factor* and is the potential energy of the magnetic moment  $\mu_I$  in the mean magnetic field  $\overline{B}_0$  of the electrons at the nucleus:

$$A = \frac{\mu_I \cdot \mu_K}{I \cdot J} \bar{B}_0. \tag{3.3}$$

The quadrupole coupling constant B is the product of the spectroscopic quadrupole moment  $Q_s$  with the gradient of the electric field  $E_z$  at the nucleus, which is produced by



Figure 3.1: The hyperfine splitting of  $^{7,9,10,11}$ Be<sup>+</sup>. All allowed dipole transitions as well as the center of gravity are shown.

the electrons:

$$B = \left(\frac{\partial E_z}{\partial z}\right)\Big|_0 \cdot eQ_s. \tag{3.4}$$

In this work only dipole transitions between electronic states with J = 1/2 are of interest. In this configuration the quadrupole contribution to the hyperfine splitting does vanish.

Since narrow linewidth lasers can drive single hyperfine transitions it is necessary to discuss two properties of the hyperfine splitting:

1. The splitting of the energy levels in different hyperfine components results in a distribution of the total transition strength in the different hyperfine transitions. The relative intensity for a hyperfine transition  $(J, I, F) \rightarrow (J', I', F')$  can be derived, according to [DRA96]:

$$S(F \to F') = \frac{(2F+1)(2F'+1)}{2I+1} \left\{ \begin{array}{cc} J' & F' & I \\ F & J & 1 \end{array} \right\}^2$$
(3.5)

where  $\{\}^2$  is the 6J symbol that relates to the coupling scheme of the hyperfine structure. For the sum of all relative intensities the sum rule

$$\sum_{F} \sum_{F'} S(F \to F') = 1 \tag{3.6}$$

holds. The hyperfine interaction does not shift the *center of gravity*, which is the energy of the unperturbed transition (compare with figure 3.1). For a spectrum with resolved hyperfine structure the center of gravity can be obtained by the following relation:

$$\nu_{cg} = \sum_{F} \sum_{F'} \nu_{F \to F'} \cdot S(F \to F'). \tag{3.7}$$

 $\nu_{F \to F'}$  is the observed transition frequency of the transition  $F \to F'$ .

2. If a transition with resolved hyperfine splitting in the lower level is excited by narrow laser radiation, a depopulation of the lower hyperfine state can occur<sup>1</sup>, because the excited electron does not have to fall back into the original hyperfine state in all cases. This has severe consequences for laser cooling and spectroscopy. To avoid the population of dark states, i.e. states that do not interact with the laser anymore, special precautions have to be considered as described in detail in section 4.3.4.

The hyperfine splitting and electric dipole transitions in the  $2s_{1/2} \rightarrow 2p_{1/2}$  resonance line of beryllium isotopes are schematically shown in figure 3.1.

#### **3.1.1** Zeeman Effect

A hyperfine level F, as described in the preceding section, has  $2F + 1 m_F$  states that are degenerated in the absence of magnetic fields. The quantum number  $m_F$  is the projection of the spin onto the z-axis that is given by, e.g. an external field. In 1886 Zeeman discovered [HAK00] in the similar case of the fine structure splitting, that this degeneration can be lifted by applying an external magnetic field, which leads to a further splitting of the hyperfine components. The theoretical calculation of the hyperfine Zeeman effect is analog to the calculation of the fine structure splitting, because the total angular momentum **F** of the atom results in a total magnetic moment  $\boldsymbol{\mu}_F$ . The potential energy of this magnetic moment in the presence of the magnetic field  $\mathbf{B} = \mathbf{B} \cdot \hat{\mathbf{e}}_z$  is given by

$$\Delta E_B = -\boldsymbol{\mu}_F \cdot \mathbf{B} \tag{3.8}$$

In the following calculation one has to differentiate between the cases of strong and weak magnetic fields. While the coupling to **F** persists in a weak magnetic field it breaks up in a strong field. In first order pertubation theory the energy splitting  $\Delta E_B$  can be derived for each  $m_F$  component based on equation 3.8:

$$\Delta E_B = g_F \cdot \mu_F \cdot m_F \cdot B, \qquad (3.9)$$

with

$$g_F = g_J \frac{F(F+1) + J(J+1) - I(I+1)}{2F(F+1)} - g_K \frac{\mu_K}{\mu_B} \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)}.$$
(3.10)

The second term can be neglected in most cases because  $\frac{\mu_K}{\mu_B} \approx 1/2000$ . From equation 3.9 it appears that the  $m_F$  components split linear to the field strength B into equidistant

<sup>&</sup>lt;sup>1</sup>optical pumping



Figure 3.2: Illustration of the energy splitting of the hyperfine levels of  $^{7,9}Be^+$  in presence of weak (Zeeman effect) and strong (Paschen Back effect) magnetic fields. The intermediate region between weak and strong magnetic field is given for spin 1/2 states by the Breit-Rabi formula (see text).

energy levels. For a better understanding of this effect figure 3.2 shows an illustration of the energy splitting in the  $2s_{1/2}$  level for <sup>9</sup>Be<sup>+</sup>. It can be calculated with the *Breit-Rabi formula*, which describes the energy splitting from weak to strong magnetic fields for  $F = I \pm \frac{1}{2}$  hyperfine states:

$$\Delta E_B \left( F = I \pm \frac{1}{2} \right) = -\frac{A}{4} + m_F g_K \mu_K B \pm \frac{\Delta E_0}{2} \left( 1 + \frac{4m_F}{2I+1} x + x^2 \right)^{\frac{1}{2}}$$
(3.11)

with

$$x \approx \frac{2\mu_B B}{\Delta E_0} \qquad \qquad \Delta E_0 = A\left(I + \frac{1}{2}\right) \qquad (3.12)$$

The graph shows the linear Zeeman region at low fields, the transition or intermediate region and the Paschen-Back region. In the Zeeman region F,  $m_F$  are still good quantum numbers, whereas the coupling between **I** and **J** breaks up in the intermediate region and F,  $m_F$  rearrange into two multiplets that are now ordered according to their  $m_J$  values since the magnetic moment of the electron is much larger than that of the nucleus.

#### **3.2** Isotope shift

The energy levels of electrons in an atom or ion are not only defined by the nuclear charge and the electromagnetic moments as they were discussed so far. The electron energy is also influenced by the finite nuclear mass and volume. While the moments cause the hyperfine splitting, nuclear mass and the finite size of the nucleus are responsible for the so-called isotope shift. The following considerations are based on works of [EWA05, SAN06b] and [OTT89].

Experiments which have been performed on Pb samples in 1919 from Merten et al. are considered to be the first ones where the isotope shift has been observed [BRE58]. It is defined as the change of an electronic transition energy between two isotopes with mass number A and A'.

$$\delta\nu^{AA'} = \delta\nu^{A'} - \delta\nu^A \tag{3.13}$$

As mentioned above this frequency change appears due to the change of the nuclear mass  $M^A$ , which is summarized in the mass effect (ME), and due to the change of the nuclear charge distribution, which is contained in the field effect (FE) and can be written as the sum of these effects

$$\delta\nu^{AA'} = \delta\nu^{AA'}_{\rm ME} + \delta\nu^{AA'}_{\rm FE} \quad . \tag{3.14}$$

**Mass Effect** The mass effect on his part is the sum of the so-called *normal mass effect* (NME) or *Bohr reduced mass correction* and the *specific mass effect* (SME) or *Hughes-Eckhart effect* [BRE58]. The former accounts for the difference in the reduced mass between two isotopes and can be calculated by

$$\delta\nu_{\rm NME}^{AA'} = \frac{(M^{A'} - M^A)}{M^A M^{A'}} \cdot m_e \cdot \nu^A = N \cdot \frac{M^{A'} - M^A}{M^A M^{A'}}$$

The latter considers all sorts of electron correlations, which also influence the motion of the nucleus. The exact calculation of this effect is extremely difficult because of mathematical difficulties with the correlation integrals. But it has the same mass dependence as the normal mass effect and can be written as:

$$\delta\nu_{\rm SME}^{AA'} = S \cdot \frac{M^{A'} - M^A}{M^A M^{A'}} \quad , \tag{3.15}$$

Hence, one can summarize the mass effect as:

$$\delta\nu_{\rm ME}^{AA'} = (N+S) \cdot \frac{M^{A'} - M^A}{M^A M^{A'}} \quad . \tag{3.16}$$

**Field Effect** The electron energy is sensitive to the charge distribution of the nucleus and can be calculated non-relativistically in first-order pertubation theory. The potential energy of the nuclear charge distribution  $\rho(\mathbf{r})$  in the electric potential of the electron shell  $V(\mathbf{r})$  can be calculated with

$$E^{A} = \int_{0}^{R} \rho(\mathbf{r}) \cdot V(\mathbf{r}) \cdot d\tau \qquad (3.17)$$

The following calculation is constrained to s electrons, which produce a spherical symmetric potential V(r) at the core. This potential can be splitted in a constant part, which represents the potential outside the nucleus and a variable part, which represents the distribution inside the nucleus, which leads to

$$E^{A} = -Ze^{2} \left\langle \frac{1}{r} \right\rangle_{\text{electron}} + \frac{2\pi}{3} Ze^{2} \left| \Psi_{\text{s}}(0) \right|^{2} \left\langle r^{2} \right\rangle_{\text{nucleus}} \quad . \tag{3.18}$$

The first term represents the interaction of one electron with a point like source with charge Ze, while the second term contains the finite size of the nucleus. It is the product of the nuclear charge density at the origin  $|\Psi_s(0)|^2$  and the radial moment of the charge distribution  $\langle r^2 \rangle_{\text{nucleus}}$  and is called *field shift (FS)*. For the isotope shift this expression becomes:

$$\delta\nu_{\rm FS}^{AA'} = E^{A'} - E^A = -\frac{2\pi}{3h} Z e^2 \Delta \left|\Psi_{\rm s}(0)\right|^2 \delta \left\langle r^2 \right\rangle_{\rm nucleus}^{AA'} , \qquad (3.19)$$

where  $\Delta |\Psi_s(0)|^2$  is the change of the electron density at the nucleus in the transition and  $\delta \langle r^2 \rangle_{\text{nucleus}}^{AA'}$  the change of the mean square charge radius. The first part can be summarized in a constant F, so that one obtains:

$$\delta\nu_{\rm FS}^{AA'} = F\delta \left\langle r_c^2 \right\rangle^{AA'} \tag{3.20}$$

Now the total isotope shift  $\delta \nu_i^{AA'}$  can be calculated by summing equation 3.16 and equation 3.20:

$$\delta\nu^{AA'} = (N+S) \cdot \frac{M^{A'} - M^A}{M^A M^{A'}} + F\delta \left\langle r^2 \right\rangle^{AA'}.$$
(3.21)

In this work the nuclear charge radii of  $^{7,10,11}$ Be are calculated based on the charge radius of <sup>9</sup>Be by measuring the isotope shift in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition, so that equation 3.21 can be remodeled to:

$$\delta \left\langle r_c^2 \right\rangle^{9,A'} = \frac{\delta \nu_{\rm IS}^{9,A'} - \delta \nu_{\rm MS}^{9,A'}}{F} = \frac{\delta \nu_{\rm FS}^{9,A'}}{F}.$$
 (3.22)

F is largely independent of the nuclear mass (compare with table 3.1) and has been calculated by Yan et al. [YAN08] to

$$F = -16.912 \ \frac{\mathrm{MHz}}{\mathrm{fm}^2}$$

and also by Puchalski et al. [PUC08] to:

$$F \approx -17.021 \frac{\text{MHz}}{\text{fm}^2},\tag{3.23}$$

with small isotopic variations as listed in table 3.1. The difference in these results is due to the inclusion of logarithmic relativistic corrections to the wave function at the origin in [PUC08]. The term  $\delta \nu_{\rm MS}^{9,A'}$  is the calculated mass shift, which will be described in detail in section 3.3. The absolute charge radii of the isotopes <sup>7,10,11</sup>Be can finally be calculated by quadratic summing:

$$r_c^{A'} = \sqrt{(r_c^9)^2 + \delta \langle r_c^2 \rangle^{9,A'}}.$$
(3.24)

#### 3.2.1 Required accuracy in this experiment

Since no experimental values for charge radii of other beryllium isotopes than <sup>9</sup>Be are available, one has to suppose that the change in the nuclear charge radii  $\delta \langle r_c^2 \rangle^{A,A'}$  is in the order of 0.5 fm<sup>2</sup> as it has been typically found in measurements of He and Li isotopes [MUE07, SAN06a]. With  $F = -16.912 \frac{\text{MHz}}{\text{fm}^2}$  and equation 3.22, the expected field shift between two Be isotopes can be roughly estimated to be about 8.5 MHz. For a substantial statement about the nuclear charge radii of <sup>7,10,11</sup>Be  $r_c^{A'}$  has to be determined with a relative accuracy comparable to that reached for the stable reference isotope <sup>9</sup>Be by electron scattering, which is approximately 1% [JAN72, BER67]. It should be mentioned that the uncertainty of the electron scattering result might be underestimated [SIC07]. The uncertainty of  $\delta \langle r_c^2 \rangle$  as calculated in equation 3.22 can be obtained by Gaussian error propagation. It is

$$\Delta\left(\delta\left\langle r_{c}^{2}\right\rangle^{9,A'}\right) = \frac{1}{F}\Delta\left(\delta\nu_{FS}^{9,A'}\right),\tag{3.25}$$

because the uncertainty of F is negligible. Therewith the uncertainty  $\Delta r_c^{A'}$  from equation 3.24 becomes:

$$\Delta r_c^{A'} = \frac{1}{2r_c^{A'}} \cdot \sqrt{\left(2 \cdot r_c^9 \cdot \Delta r_c^9\right)^2 + \left(\frac{1}{F}\Delta\left(\delta\nu_{FS}^{9,A'}\right)\right)^2}.$$
(3.26)

Using these two equations, it can be estimated that the required accuracy in the determination of the field shifts is approximately 2 MHz, e.g., the accuracy of the experimentally determined isotope shift **and** the calculated mass shift contribution should be at least in the order of 1 MHz.

### 3.3 Mass effect calculations in three electron systems

This section is based on a presentation of G. Drake at GSI in January 2005 [DRA05] and correlated works [DRA95, DRA99]. As already mentioned, the calculation of the specific mass effect is difficult due to the electron correlations. A recipe for the calculation of the mass effect in two and three-electron systems and the extraction of nuclear charge radii can be given as follows:

- 1. Calculation of wave functions and eigenvalues of the non-relativistic Schrödinger equation for lithium-like ions to spectroscopic accuracy.
- 2. Inclusion of the finite mass (and mass polarization) up to second-order pertubation theory.
- 3. Inclusion of relativistic and QED effects.
- 4. Comparison of the calculated energies with high precision experiments.
- 5. Extraction of the nuclear volume effect and hence the nuclear charge radius from the difference between the experimentally determined isotope shift and the theoretically calculated mass shift.

Table 3.1: Calculated mass shifts for <sup>7,10,11</sup>Be<sup>+</sup> relative to <sup>9</sup>Be<sup>+</sup> for the D1 and D2 transitions. Values from [YAN08] are corrected for improved mass measurements since the publication. Hence, the same masses were used as in [PUC08]. The remaining difference in the mass shift of <sup>11</sup>Be is caused by inclusion of the nuclear polarization effect of 208 kHz in [PUC08].

Isotope	$2s_{1/2} \rightarrow 2p_{1/2}$	$2s_{1/2} \rightarrow 2p_{3/2}$	С	Reference
	MHz	MHz	$\rm MHz/fm^2$	
$^{7}$ Be	-49 225.780(39)	-49 231.84(4)	-16.921	[YAN08]
	$-49\ 225.736(35)$		-17.021(31)	[PUC08]
$^{10}\mathbf{Be}$	$17 \ 310.442(13)$	$17\ 312.57(6)$	-16.921	[YAN08]
	$17\ 310.437(13)$		-17.027(31)	[PUC08]
$^{11}\mathbf{Be}$	$31 \ 560.087(24)$	31 563.96(2)	-16.921	[YAN08]
	$31 \ 560.302(31)$		-17.020(31)	[PUC08]

The calculation of non-relativistic wave functions for two- and three-electron systems is best performed by introducing Hylleraas' coordinates, which treat correlations of the electrons in an efficient way [HYL28]. With these coordinates it is possible to calculate most of the correlation energy already with a relative small set of basis states. A continuous improvement of this method permitted for example an enhancement of the relative accuracy of the non relativistic ground state energy for He up to  $10^{-19}$  [DRA99].

A Generalization to three-electron systems was hampered by the fact that the occurring correlation integrals do not have closed solutions and that the numerical integration converges slowly. Developments of Drake and Yan increased the speed of the needed numerical integration by 2-3 orders of magnitude [DRA95]. This development was the starting point for high precision calculations for Li [YAN00, YAN03, PUC06] and Be<sup>+</sup> [YAN08, PUC08]. The following section summarizes the mass shift calculation in a threeelectron system.

The total binding energy  $E_{\text{tot}}$  for an electron can be calculated by

$$E_{\rm tot} = E_{\rm NR} + \alpha^2 E_{\rm rel} + \alpha^3 E_{\rm QED} + \dots + \Delta E_{\rm FE}$$
(3.27)

with the non-relativistic energy  $E_{\rm NR}$ , the relativistic and QED corrections  $E_{\rm rel}$  and  $E_{\rm QED}$ and the contribution  $\Delta E_{\rm FE}$  of the finite size of the nucleus. Introducing the expansion for mass polarization, one obtains

$$E_{\rm NR} = E_{\rm NR}^{(0)} + \frac{\mu}{M} E_{\rm NR}^{(1)} + \left(\frac{\mu}{M}\right)^2 E_{\rm NR}^{(2)} + \dots$$
(3.28)

$$E_{\rm rel} = E_{\rm rel}^{(0)} + \frac{\mu}{M} E_{\rm rel}^{(1)} + \left(\frac{\mu}{M}\right)^2 E_{\rm rel}^{(2)} + \dots$$
(3.29)

$$E_{\text{QED}} = E_{\text{QED}}^{(0)} + \frac{\mu}{M} E_{\text{QED}}^{(1)} + \left(\frac{\mu}{M}\right)^2 E_{\text{QED}}^{(2)} + \dots$$
(3.30)

The first step for further calculations is the determination of non-relativistic wave functions, which satisfy the variational condition

$$\delta \int \Psi(\hat{H}_0 - E) \Psi d\tau = 0. \tag{3.31}$$

The Hamiltonian  $\hat{H}_0$  for three-electron systems is the sum of the kinetic energy of each electron, the Coulomb energy for each electron in presence of an infinite heavy core with charge Z and the Coulomb energy of each electron in presence of the other electrons. It is

$$\hat{H}_0 = -\frac{1}{2} \sum_{i=1}^3 \nabla_i^2 - Z \sum_{i=1}^3 \frac{1}{r_i} + \sum_{i>1}^3 \frac{1}{r_{ij}}$$
(3.32)

Introducing the correlation of the electrons,  $\rho_i \rho_j$ , an additional part has to be added which leads to the total Hamiltonian  $\hat{H}$ 

$$\hat{H} = \hat{H}_0 + \lambda \hat{H}', \qquad (3.33)$$

with the additional Hamiltonian  $\hat{H}'$  which is given by

$$\hat{H}' = \sum_{i>j}^{3} \nabla_i \nabla_j. \tag{3.34}$$

These equations are given in atomic units and a rescaling of the distance r

$$r \to \frac{m_e}{\mu} r \tag{3.35}$$

and energy E

$$E \to E \frac{\hbar^2}{e^4 \mu} \tag{3.36}$$

has been performed.  $\mu$  is the reduced mass of the three-electron system and is given by

$$\mu = \frac{M}{\frac{M}{m_e} - 2}.\tag{3.37}$$

For  $\hat{H}_0$  it is possible to calculate the wave function  $\Psi_0$  by variational calculations, where the number of basis states are gradually increased and finally extrapolated into the full Hilbert space. Thus, the wave function and the binding energy of the electron are determined.  $\hat{H}_0$  and its eigenvalue describe the normal mass shift, while  $\hat{H}'$  depicts the specific mass effect. As the solution for  $\hat{H}_0$  can be determined,

$$\hat{H}\Psi = E\Psi \tag{3.38}$$

can be solved with time independent pertubation theory, where  $\lambda$  is the pertubation coefficient. Wavefunction and energy can now be written as

$$\Psi = \Psi_0 + \lambda \Psi_1 + \lambda^2 \Psi_2 + \dots \tag{3.39}$$

$$E = E_0 + \lambda E_1 + \lambda^2 E_2 + \dots (3.40)$$

With the obtained wave function  $\Psi$  for the non-relativistic case at finite mass of the nucleus with charge Z it is possible to calculate relativistic and QED corrections up to the order of  $\frac{\mu}{M}\alpha^5mc^2$ . The shift between the binding energy for different isotopes can be easily calculated by the difference of equation 3.27 for different isotopes, where all mass-independent contributions cancel out. This is of particular importance, since the mass independent QED correction in first-order has a relatively large uncertainty that would not allow to determine the theoretical mass shift with the required accuracy. The final result for the isotope shift is:

$$\Delta E^{AA'} = \left[ \left( \frac{\mu}{M} \right)_{A} - \left( \frac{\mu}{M} \right)_{A'} \right] \left( E^{(1)}_{NR} + \alpha^2 E^{(1)}_{rel} + \alpha^3 E^{(1)}_{QED} \right)$$
(3.41)

$$+\left[\left(\frac{\mu}{M}\right)_{A}^{2}-\left(\frac{\mu}{M}\right)_{A'}^{2}\right]\left(E_{NR}^{(2)}+\alpha^{2}E_{rel}^{(2)}+\alpha^{3}E_{QED}^{(2)}\right)$$
(3.42)

$$+\dots \qquad (3.43)$$

$$+\underbrace{\Delta E_{FE}^{A} - \Delta E_{FE}^{A'}}_{\text{Field shift}} \tag{3.44}$$

The calculated mass shifts for different Be<sup>+</sup> isotopes relative to <sup>9</sup>Be<sup>+</sup> in the  $2s_{1/2} \rightarrow 2p_{1/2}$  and  $2s_{1/2} \rightarrow 2p_{3/2}$  transitions are summarized in table 3.1. In [PUC08] also core polarization effects, the excitation of the core in presence of the electric field of the electron, are included, which lead to the difference of the mass shift in <sup>11</sup>Be<sup>+</sup> compared to [YAN08]. The agreement between the independent theoretical calculations is excellent and the uncertainty of a few kHz is much better than required according to section 3.2.1. Finally it should be mentioned that the total transition frequency of the D1 line in <sup>9</sup>Be<sup>+</sup> has also been calculated by [PUC08] to

$$\nu^{\text{theory}} = 31928.734(8) \frac{1}{\text{cm}} \tag{3.45}$$

and by [YAN08] to

$$\nu^{\text{theory}} = 31928.738(5) \frac{1}{\text{cm}}.$$
(3.46)

## Chapter 4

## Laser spectroscopy on ions in traps

In 1953 W. Paul worked on the idea that trajectories of molecules and particles with a dipole moment can be focused by electric or magnetic multipole fields. After the development of lenses for atom and molecule beams the idea was expanded to charged particles. The interaction of charged particles with an electrodynamic field permitted the confinement of ions in two dimensions and led to the development of quadrupole mass filters [PAU53, PAU55, PAU58]. Until 1959 W. Paul and his co-workers produced the first three dimensional cylindrical ion trap [FIS59]. Both, the development of quadrupole mass spectrometers and the development of ion traps were epoch-making episodes in physics. They quickly found their applications in analytical chemistry, biology, geology and nowadays in precision experiments and quantum logic. As honour for his pioneering work in the field of ion confinement W. Paul received the Nobel-price in physics in 1989. Prestage, Dick and Malecki proposed the linear Paul trap as a minor variation of the quadrupole mass spectrometer [PRE89]. As an advantage the authors mainly claimed the higher storing capacity compared to cylindrical Paul traps and the good optical accessibility.

The ultimate sample for atomic spectroscopy is a cold cloud of atoms or ions or even single particles at rest. Thanks to the development of ion traps such experiments became feasible, where all sources of additional line broadening can be avoided in order to study transitions at the resolution limit given by their natural linewidth. In 1980 the first single Ba<sup>+</sup> ion was stored and cooled in a Paul trap [NEU80] and the first single Mg<sup>+</sup> ion was stored in a Penning trap in 1980 [WIN81]. For the reduction of the ion's kinetic energy in a trap some possibilities exist. For a Paul trap buffer gas cooling or, if an appropriate excitation scheme exists, laser cooling can be used. With buffer gas cooling the kinetic energy of the ions can only be reduced to room temperature (about 1 eV), while it can be decreased below 1 meV by laser cooling, which is sufficient for most experiments. The combination of ion and atom traps with cooling techniques initiated a wide variety of high resolution experiments in many fields of physics, like e.g. precision tests of fundamental symmetries at low energy or nuclear structure investigation.

In this chapter theoretical calculations for ion confinement in two and three dimensions will be explained. It ends with an overview about the principles of laser cooling.



Figure 4.1: Hyperbolic electrodes (left) and the corresponding quadrupole potential of a Paul filter (right).

### 4.1 Quadrupole mass filter

An ion is bound harmonically at a point in space if a force acts on the ion, that increases proportional to its distance to the point. Hence, the potential  $\Phi(x, y, z, t)$  has to have the form

$$\Phi(x, y, z, t) = \Phi_0(t) \cdot \left(\alpha x^2 + \beta y^2 + \gamma z^2\right).$$
(4.1)

Neglecting space charge effects and induced magnetic fields, equation 4.1 has to fulfill the Laplace equation  $\Delta \Phi = 0$ , which is only possible for

$$\alpha + \beta + \gamma = 0. \tag{4.2}$$

Two intuitive possibilities to meet this demand are

- 1.  $\alpha = -\beta$  and  $\gamma = 0$ : expresses the two dimensional confinement as it is used e.g. for mass filter.
- 2.  $\alpha = \beta$  and  $\gamma = 2\alpha$ : expresses the three dimensional confinement as it is used for ion traps.

Even if the Paul trap is not used as a mass filter, it is helpful to describe the quadrupole mass filter first for a better understanding of the ion's motion in the trap. Inserting  $\alpha = -\beta = 1/r_0^2$  into equation 4.1 leads to a two dimensional quadrupole field:

$$\Phi(x, y, t) = \Phi_0(t) \cdot \frac{x^2 - y^2}{r_0^2}.$$
(4.3)

This field can be produced by four hyperbolic electrodes as shown in figure 4.1 or, more commonly, by round rods, which should fulfill the condition

$$\frac{r_{\rm rod}}{r_0} = 1.14511\tag{4.4}$$

for the ratio between field free radius  $r_0$  and rod radius  $r_{\rm rod}$  [LEE71] in order to have a good approximation of a quadrupole field. Opposed electrodes have the same potential, while neighboring electrodes have opposite sign, but the same amplitude (compare with figure 4.1). Applying now a radio frequency (rf) field

$$\Phi_0(t) = V \cdot \cos(\Omega t) \tag{4.5}$$

with oscillation period  $T = \frac{2\pi}{\Omega}$  and amplitude V, confines ions, propagating along the z axis. For t = 0 they will be focused into x direction and defocused in y direction, but for t = T/2 they will be focused in y direction and defocused into x direction or the other way around.

#### 4.1.1 Equations of motion in a mass filter

The mass selective properties of quadrupole mass filters can be obtained by adding a constant DC voltage U to  $\Phi_0(t)$ . It converts equation 4.3 to:

$$\Phi(x, y, t) = [U + V \cdot \cos(\Omega t)] \cdot \frac{x^2 - y^2}{r_0^2}.$$
(4.6)

The equations of motion for all three spatial directions are given by:

$$\ddot{x} + \frac{2e}{mr_0^2} \left[ U + V \cdot \cos(\Omega t) \right] \cdot x = 0$$
(4.7)

$$\ddot{y} + \frac{2e}{mr_0^2} \left[ U + V \cdot \cos(\Omega t) \right] \cdot y = 0$$
(4.8)

 $\ddot{z}$ 

$$= 0$$
 (4.9)

This system of decoupled differential equations can be solved using a somehow artificial substitution. With

$$\omega t = 2\zeta \qquad a_u = a_x = -a_y = \frac{8eU}{mr_0^2 \Omega^2} \qquad q_u = q_x = -q_y = \frac{4eU}{mr_0^2 \Omega^2} \qquad (4.10)$$

equation 4.7 and 4.8 are transformed into the normal form of Mathieu's differential equation with u = x, y [MEI54].

$$\frac{d^2u}{d\zeta^2} + \left[a_u - 2q_u \cdot \cos 2\zeta\right] \cdot u = 0 \tag{4.11}$$

Solutions for this equation can be expressed in terms of a linear combination of two infinite Fourier series. They are usually denoted as

$$u(\zeta) = A \cdot e^{\mu_u \zeta} \sum c_{2s,u} e^{2is\zeta} + B \cdot e^{-\mu_u \zeta} \sum c_{2s,u} e^{-2is\zeta}.$$
 (4.12)

The start parameters u(0),  $\dot{u}(0)$  determine the coefficients A and B, while  $c_{2s,u}$  and  $\mu_u$  are only depending on  $a_u$  and  $q_u$ .  $\mu_u$  is called the characteristic exponent, which defines the kind of solution for equation 4.12. Four possibilities exist:



Figure 4.2: Stability diagram of a mass filter or a linear Paul trap for x and y direction up to stability region III (left) and an enlarged view ofstability region I for which stable trajectories in x and y direction occur simultaneously.

- 1.  $\mu_u$  is real; the solutions grow uncontrolled and are unstable
- 2.  $\mu_u$  is complex; for some start parameters the solutions are stable
- 3.  $\mu_u = i\beta_u$  and  $\beta_u$  is no integer ; the solutions are periodic and stable
- 4.  $\mu_u = i\beta_u$  and  $\beta_u$  is integer; the solutions are at the border between stable and unstable solutions

A plot in which stable solutions for equation 4.12 are shown in dependence of  $q_u$  on the abscissa and  $a_u$  on the ordinate is called stability diagram and illustrated in figure 4.2. To describe an ion's stable trajectory through the mass filter one has to consider the intersection between the stability plot for x and y axis. Stability domain I is also shown enlarged in figure 4.2.

A quadrupole mass filter is operated that way, that DC voltage U and RF voltage V are varied in a constant proportion of a/q = 2U/V. This ratio can be opted that way, that the load line, which starts at (0,0) with the slope a/q, cuts the stability diagram, which allows the stable transmission of a mass interval, which is clearly defined through the interval  $\Delta q = \frac{4e\Delta V}{\Delta m r_0^2 \Omega^2}$ . The best resolution for a mass spectrometer would theoretically be achieved by choosing the ratio a/q so, that it cuts the stability diagram at the peak  $(a_0 = 0.237, q_0 = 0.706)$ . This  $\delta$  function transmission peak can not be reached in an experiment, due to fringe fields, geometry errors and other inadequacies. For U = 0, in the rf-only mode,  $\Delta q$  becomes maximal and the filter acts as a high pass filter and all masses above  $m_{\min} = \frac{4eV}{0.92r_0^2\Omega^2}$  will be transmitted. For  $a, q \ll 1$ , the so called adiabatic approximation, stable solutions for x(t) and y(t) can be given by:

$$x(t) = x_0 \cos(\omega_x t + \varphi_x) \left(1 + \frac{q_x}{2} \cos \Omega t\right)$$
(4.13)

$$y(t) = y_0 \cos(\omega_y t + \varphi_y) \left(1 + \frac{q_y}{2} \cos \Omega t\right)$$
(4.14)

 $x_0, \varphi_x$  and  $y_0, \varphi_y$  are the initial values, while  $\omega_x$  and  $\omega_y$  are:

$$\omega_x = \frac{\Omega}{2} \sqrt{\frac{q_x^2}{2} + a_x} \tag{4.15}$$

$$\omega_y = \frac{\Omega}{2} \sqrt{\frac{q_y^2}{2}} + a_y \tag{4.16}$$

That means the trajectory in x and y direction is comparable to a harmonic oscillation  $\omega_{x,y}$  (secular motion) with an superimposed oscillation of frequency  $\Omega$  (micro motion), which is the driving frequency of the RF field. Neglecting the micromotion the solution is equal to the particle motion in a pseudo potential  $\Psi$  of the form:

$$\Psi = \frac{m}{2} \left( \omega_x^2 x^2 + \omega_y^2 y^2 \right) \tag{4.17}$$

The depth of this potential can be calculated according to [MAJ05]:

$$\Psi_x = \frac{m}{2}\omega_x^2 r_0^2 \tag{4.18}$$

$$\Psi_y = \frac{m}{2} \omega_y^2 r_0^2 \tag{4.19}$$

### 4.2 Linear Paul trap

To transform a mass filter, as described in section 4.1, into a linear trap the quadrupole potential  $\Phi(x, y, t)$  has to be superimposed with a positive (for positive ions), constant potential in z direction  $\Phi(z)$ . In order to have a trapping potential that is well defined along the z axis it should have a parabolic shape with its minimum in the middle of the trap. Three main possibilities to achieve the needed potential shape exist and are shown in figure 4.3.

- a Segmentation of all four electrodes into at least 3 parts. All electrodes carry the RF potential, while the end electrodes carry an additional DC potential.
- b Segmentation of all four electrodes into at least 3 parts. Only the middle electrodes carry the RF potential, while the DC potential is applied to the end electrodes.
- c Segmentation of two opposing electrodes into at least 3 parts. The segmented electrodes carry the DC potential, while the unsegmented electrodes carry the RF potential.



Figure 4.3: Three main possibilities exist to modify a quadrupole mass filter into a linear Paul trap. Top and side view illustrate the needed potentials to achieve trapping conditions. Further information can be found in the text.

It is obvious that there is no way to create a potential  $\Phi(z)$ , that is really independent of the x and y coordinate. The following description constrains to the case of ion motion near the trap axis in the middle of the trap, which will justify this approach. With these restrictions case 2 and 3 can be treated the same way and case 1 can be understood as case 2, where a DC offset  $U_{\text{offset}} = V$  is added to all electrodes.

Referring to the most general case 2 the quadrupole potential  $\Phi(x, y, z, t)$  along the whole trap length becomes in presence of the static trapping potential:

$$\Phi(x, y, z, t) = \left[\frac{U_z}{z_0^2} + \Phi_0(t)\right] \cdot \left(\alpha x^2 + \beta y^2 + \gamma z^2\right).$$
(4.20)

To solve the Laplace equation in this case one has to consider the case in which  $\alpha = \beta = 1/2$ , with  $\gamma = 1$ . Now the quadrupole potential becomes

$$\Phi(x, y, z, t) = \left[U + V \cdot \cos(wt)\right] \cdot \frac{x^2 - y^2}{r_0^2} + \frac{U_z}{z_0^2} \cdot \left[z^2 - \frac{x^2 + y^2}{2}\right].$$
 (4.21)

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As a result the equations of motion change now to:

$$\ddot{x} + \frac{2e}{mr_0^2} \left[ U + V \cdot \cos(wt) \right] \cdot x - \frac{eU_z}{mz_0^2} \cdot x = 0$$

$$(4.22)$$

$$\ddot{y} + \frac{2e}{mr_0^2} \left[ U + V \cdot \cos(wt) \right] \cdot y - \frac{eU_z}{mz_0^2} \cdot y = 0$$

$$(4.23)$$

$$\ddot{z} + \frac{2eU_z}{mz_0^2} \cdot z = 0 \tag{4.24}$$

Using the normal form of Mathieu's differential equation for this case and an additional substitution of

$$b = \frac{8eU_z}{m\omega^2 z_0^2} \tag{4.25}$$

the equations of motion change to:

$$\frac{d^2x}{d\zeta^2} + \left[a - \frac{b}{2} - 2q \cdot \cos 2\zeta\right] \cdot x = 0$$
(4.26)

$$\frac{d^2y}{d\zeta^2} + \left[a + \frac{b}{2} - 2q \cdot \cos 2\zeta\right] \cdot y = 0 \tag{4.27}$$

$$\frac{d^2z}{d\zeta^2} + b \cdot z = 0 \tag{4.28}$$

Also in this case the movement along the z axis is independent of the radial movement. The solution is a harmonic oscillation

$$z(t) = Z_1 e^{i\omega_z t} + Z_2 e^{-i\omega_z t}$$
(4.29)

with frequency

$$\omega_z = \frac{\sqrt{b}\omega}{2} = \sqrt{\frac{2eU_z}{mz_0^2}}.$$
(4.30)

For the adiabatic approach the pseudo-potential in radial direction can be derived analogous to equation 4.18 and 4.19, while the depth of the pseudo-potential in z direction can be calculated in first order by:

$$\Psi_y = k \cdot q \cdot U \tag{4.31}$$

k is called geometry factor, which is approximately 0.025, q the charge of the trapped ion and U the applied DC potential [MAJ05].

### 4.3 Theory of laser cooling

It is possible to allocate a momentum  $\mathbf{p} = \hbar \mathbf{k}$  to mass less particles as e.g. photons. This momentum can be devolved to baryonic particles and as sufficient powerful lasers are nowadays available, this effect can be used in many ways. The following review about laser cooling is based on [MET99] and will be complemented with other publications from [MIT82, MIN86, BAL85, SCH01, HOE05].

#### 4.3.1 Saturation intensity

A two level system (atom or ion) with a ground state population  $N_1$  and an excited state population  $N_2$  has the total population  $N_1 + N_2 = 1$ . The decay rate from  $N_2$  to  $N_1$  is defined by  $\Gamma = \frac{1}{\tau}$ , with the lifetime  $\tau$  of the excited state. If the system is irradiated with a laser of Intensity I and the laser frequency is equal to the resonance frequency the saturation parameter is defined as:

$$s = 2\left(\frac{\Omega}{\Gamma}\right)^2 = \frac{I}{I_s}.$$
(4.32)

 $\Omega$  is the Rabi frequency and  $I_s$  the so-called saturation intensity. If the irradiated laser has an intensity of

$$I_s = \frac{\pi h c}{3\lambda^3 \tau} \tag{4.33}$$

the transition experiences a power broadening of  $\sqrt{2}$  and the population of the excited state is 1/4. For Be<sup>+</sup> the saturation intensity in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition is

$$I_s = 1.3 \frac{\text{mW}}{\text{mm}^2}.$$
 (4.34)

#### 4.3.2 Scattering rate

A two level system as described above scatters photons when the transition is probed by a laser beam. Each photon transfers its momentum to the system. The system absorbs photons which come from the same direction, but during the emission process they are randomly emitted in  $4\pi$ . The result is a net momentum transfer in direction of the laser beam. This process is called radiation pressure and is the principle of laser cooling. The effective scattering rate  $\gamma$  is determined by the frequency detuning  $\delta$  between laser and resonance frequency, the Doppler shift  $\omega_D$ , the laser intensity and the maximum scattering rate  $\Gamma$ . It is:

$$\gamma = \frac{s\Gamma/2}{1+s+(2(\delta+\omega_D)/\Gamma)^2}.$$
(4.35)

#### 4.3.3 Spontaneous scattering force

A two level system with resonance frequency  $\omega_0$  is irradiated with a laser of frequency  $\omega$  and intensity *I*. The linewidth of the laser is assumed to be small compared to the natural linewidth of the transition. The detuning between laser and resonance frequency  $\delta$  is defined by  $\delta = \omega - \omega_0$ . A system that is propagating with velocity v in direction (+) or opposing direction (-) of the laser beam experiences a laser field with a frequency shift  $\delta$  due to the optical Doppler shift that is

$$\delta = \omega - \omega_0 \pm kv. \tag{4.36}$$

The momentum that is carried by one photon of light with wavelength  $\lambda$  is

$$p = \hbar k. \tag{4.37}$$
The average force, the *spontaneous scattering force*, that affects the system can now be derived by the photon's momentum times the averaged scattering rate. It is

$$F_{\pm} = \pm \hbar k \cdot \frac{s\Gamma/2}{1 + s + (2(\delta \pm kv)/\Gamma)^2}.$$
(4.38)

The momentum transfer of photons to ions and atoms found many applications in the last years. It is needed for all experiments dealing with quantum logic, Bose Einstein condensates, frequency standards and also in ultra trace analysis [MON95, PER02, BER98, HOE05]. In regard to trapped ions it is used for the reduction of their kinetic energy. An ion, caught in a trap, is oscillating in all three spatial directions. Photons of a red detuned laser, which is applied along the trap axis, will be absorbed by ions that move in opposite direction compared to the laser and will lose kinetic energy in this direction as soon as they get into resonance. If several ions are trapped the kinetic energy of all spatial directions will be reduced with a single cooling laser, because the equations of motion will be coupled in this case due to Coulomb interaction between the ions.

#### 4.3.4 Doppler limit

The minimal kinetic energy that can be reached by the spontaneous scattering force is called Doppler limit. It is obvious, that the atom cannot dissipate all its energy in presence of the laser field, because the continuos absorption and emission process results in a vibration of the ion and can be expressed in terms of the temperature  $T_D$ .

$$T_D = \frac{\hbar}{2} \frac{\Gamma}{k_B} \tag{4.39}$$

For Be<sup>+</sup> it is approximately  $T_D = 80 \ \mu$ K. The expected Doppler broadening at this temperature is around 2 MHz. When the cooling laser is shined in from one side only the ions experience a net force that pushes the ions in direction of the laser field, but the axial trapping potential is strong enough to keep the ions stored.

#### 4.3.5 Cooling transition

For an efficient laser cooling process high scattering rates and closed two level systems are required. The scattering rate which is defined by the lifetime of the excited state is with  $\tau \approx 9 \ ns$  sufficiently high for Be<sup>+</sup>. But due to the nuclear spin the energy levels of the isotopes <sup>7,9,11</sup>Be<sup>+</sup> will split according to the hyperfine splitting as illustrated in figure 3.1, which allows optical pumping into dark states. The following considerations restrict to the case of <sup>9</sup>Be<sup>+</sup> that is the only available isotope for off-line tests - they can be easily assigned to <sup>7,10,11</sup>Be<sup>+</sup>.

The energy splitting of the ground state with 1.25 GHz is small compared to room temperature, so that it is reasonable to assume that both ground states are equally populated. The branching ratios show that an ion, that is excited from the F = 1 state has a probability of 70 % to be in the F = 1 state after emission or 60 % if the ion is excited from the F = 2 state [SCH01], which would require a repumping laser with a frequency detuning of 1.25 GHz and approximately the same intensity.

In experiments which use laser cooled <sup>9</sup>Be<sup>+</sup> ions for sympathetic cooling of light molecules [SCH01, ROT09] a different approach is used to ensure high laser cooling efficiency. They use the  $2s_{1/2}(F = 2) \rightarrow 2p_{3/2}$  transition, which has a very small hyperfine splitting of 8.9 MHz in the  $2p_{3/2}$  state, which cannot be resolved due to the natural linewidth. When a small magnetic field of several Gauss is applied along the trap axis the degeneration of the  $m_{\rm F}$  components in the  $2p_{3/2}(F = 0, 1, 2, 3)$ states is nullified. When the cooling laser is  $\sigma^{\pm}$  polarized optical pumping appears, which leads due to selection rules theoreti-



Figure 4.4: Illustration of the  $2s_{1/2}$  and  $2p_{3/2}$  substates in presence of a magnetic field. If  $\sigma^{\pm}$  polarized light is used, optical pumping into the  $2s(F = 2, m_F = \pm 2)$ occurs. The resective  $2s(F = 2, m_F = \pm 2) \rightarrow 2p(F =$  $3, m_F = \pm 3)$  transitions are closed two-level systems which can be used as cooling transitions of trapped ions.

cally to the closed two level system  $2s(F = 2, m_F = \pm 2) \rightarrow 2p(F = 3, m_F = \pm 3)$  as illustrated in figure 4.4. But also the optical pumping process is disturbed by residual magnetic fields and other effects which make it necessary to use a repumping laser. But instead of the relatively high power of the repumping laser without optical pumping only a few percent are needed in this case. Instead of using a second laser, an EOM<sup>1</sup> is used to generate side bands with a distance of 1.25 GHz to the carrier frequency. This approach is not suitable for laser cooling of different Be isotopes, because the hyperfine splitting of the ground state spreads a range of 1.2 GHz to more than 2.7 GHz for all Be isotopes. Microwave amplifiers, which cover such a big frequency range with the required power of more than 10 W, which is needed for non resonant EOM devices, are not available. This means, that the experimental setup during an on-line beam time would have to be changed, which is undesirable.

Another possibility, that can be used and will be tested in this work is the mixing of the ground states F = 1, 2 with directly irradiated microwaves. In this case any of the hyperfine transitions in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition can be used, if the transition probability is not too small and EOMs or magnetic coils do not have to be used at all. In this work the  $2s_{1/2}(F = 1) \rightarrow 2p_{3/2}(F = 2)$  transition will be used for laser cooling. Trapped Be ions will be irradiated with the cooling laser that is shined into the trap along the trap axis under a slight angle. The laser frequency is shifted from far below the resonance frequency (5 GHz) in approximately 5 sec to  $\Gamma/2$  below the resonance which are experience values in other working groups [WER09, ROT07]. The far detuning of the cooling laser will initially cool the faster ions inside the trap, which dramatically reduces the needed cooling time. The laser beam diameter is approximately 1 mm to irradiate

 $<sup>^{1}</sup>$ Electro optical modulator



Figure 4.5: Measuring scheme for alternating cooling and spectroscopy of the ions based on the technique used in [WAN04]. For further details see text.



Figure 4.6: Measuring scheme based on a chain of crystalized Be<sup>+</sup> ions as used in [HER09].

the whole capture volume with a laser power of 5 to 10 mW, which is much higher than the saturation intensity of 1.3  $\frac{mW}{mm^2}$ .

## 4.4 Spectroscopy of laser cooled Be<sup>+</sup> ions

When the caught ions have been cooled down to the Doppler limit and the phase transition to an ion crystal is reached spectroscopy can be performed. Unfortunately it is not possible to do a full frequency scan over the hyperfine structure components with the cooling laser, because the ions will be heated up immediately when the frequency gets close to the resonance of the examined transition. To avoid this heating two possibilities exist which have to be investigated in off-line tests in regard to their practicability.

### 4.4.1 Switching between cooling and spectroscopy laser

One possibility, which is illustrated in figure 4.5, is the fast switching between cooling and spectroscopy laser as it was successfully used in isotope shift measurements of He [WAN04]. While the cooling laser has a fixed frequency, which is about  $\Gamma/2$  below the resonance frequency, the spectroscopy laser is continuously scanned in a frequency range of approximately 20 MHz around the investigated hyperfine transition. When the cooling laser is shined into the trap the spectroscopy laser is blocked. After approximately 8  $\mu$ s the cooling laser is blocked and the spectroscopy laser is shined in for approximately 2  $\mu$ s and the fluorescence light is detected with a photo multiplier and the laser frequency at this moment is recorded. This procedure is repeated several times. Whenever the ions are heated up due to a blue shifted frequency of the spectroscopy laser they will be cooled down again and the next cycle begins. As a result of the continuous scanning of the spectroscopy laser the fluorescence light will be recorded at random frequencies and line shifts due to the velocity change of the ion during spectroscopy will become negligible.

## 4.4.2 Spectroscopy in the ion crystal

Another possibility is spectroscopy on single ions in an ion crystal as illustrated in 4.6. This technique has recently been used for spectroscopy of Mg<sup>+</sup> in a linear Paul trap [HER09]. In this experiment a Mg ion chain is created. The cooling laser is applied under an angle, so that only one side of the ion chain is illuminated with the cooling laser. The other side is sympathetically cooled by Coulomb interaction between the ions. The spectroscopy laser has approximately  $\frac{1}{10}$  of the cooling laser power and is shined into the trap in axial direction. The fluorescence light of an ion at the side, which is not illuminated with the cooling laser is recorded with a spatial resolved photomultiplier.

The biggest advantage of this setup, which could be easily adapted for spectroscopy on Be is that the ion crystal could consist of <sup>10</sup>Be during on-line measurements, which is most comfortable for laser cooling. All other isotopes which would be loaded additionally into the trap would be separated by the light pressure of the cooling laser, so that also here spatially resolved spectroscopy is possible.

## Chapter 5

## Laser spectroscopy on fast atoms and ions

The spectroscopy of cooled ions or atoms in a trap as described in the previous chapter provides the ultimate resolution for laser spectroscopy. However, for most studies of shortlived isotopes on-line such a high resolution is not required. Moreover, very short-lived isotopes with lifetimes below 100 ms can hardly be addressed with trap measurements. For these cases, S.L. Kaufmann suggested in 1976 a technique which provides relatively high-resolution and is at the same time very general and applicable even for isotopes with lifetimes of a few ms [KAU76]. The idea was to use the longitudinal phase-space compression in the acceleration of ions in a conservative field by performing collinear laser spectroscopy on a fast atom or ion beam. This proposal was realized soon and its feasibility was demonstrated in experiments. The principle and limitations of this technique will be briefly discussed in this chapter.

## 5.1 The principle of collinear laser spectroscopy

Considering an ion source at a temperature T, the thermal distribution of ion velocities is expected to have a Maxwell-Boltzmann shape with the most probable velocity

$$\alpha = \langle \upsilon \rangle = \sqrt{\frac{2k_BT}{m}}.$$
(5.1)

This leads to a Doppler broadening of the resonance line (rest-frame frequency  $\nu_0$ ) with a full width half maximum (FWHM)

$$\Gamma_D = \sqrt{\frac{2k_B T}{mc^2 \ln 2}} \nu_0 = 7.162 \times 10^{-7} \sqrt{\frac{T}{M} \left[\frac{K}{\text{amu}}\right]} \nu_0.$$
 (5.2)

This results in a linewidth of approximately 10 GHz for the resonance line of <sup>9</sup>Be at 2000 K, which is almost as large as the complete isotope shift between <sup>7</sup>Be and <sup>9</sup>Be. Hence, laser spectroscopy in the hot production and ionization region of an on-line source can only be used for very heavy elements since the Doppler width reduces with  $1/\sqrt{M}$  and

the nuclear size effect increases with  $Z^2$ . This approach has recently been used, e.g., for polonium [COC08] and lead [SEL06] at the resonance ionization laser ion source (RILIS) at ISOLDE.

The velocity distribution of the ions can be reduced if they are accelerated to kinetic energies of a few 10 keV in an electrostatic potential. Since all ions experience the same increase in energy, independent of their start velocity, the energy width  $\delta E$  of the ensemble stays constant. As an example, consider the kinetic energy distribution in the ion source may be given by velocities ranging from  $v_1 = 0$  to  $v_2 = \alpha$ . After acceleration through the voltage difference U the kinetic energy of the ions is in the range

$$E_1 = eU, \quad E_2 = \frac{1}{2}m\alpha^2 + eU$$
 (5.3)

and the velocity spread is now given by

$$v_1' = \sqrt{\frac{2eU}{m}}, \quad v_2' = \sqrt{\alpha^2 + \frac{2eU}{m}}.$$
 (5.4)

The latter can be expanded to

$$v_{2}' = \sqrt{\frac{2eU}{m}} \sqrt{\frac{m\alpha^{2}}{2eU} + 1} \approx v_{1}' \left( 1 + \frac{1}{2} \frac{k_{B}T}{eU} \right) = v_{1}' \left( 1 + \frac{\alpha^{2}}{2v_{1}'^{2}} \right), \tag{5.5}$$

and, hence, the velocity difference is

$$\upsilon_2' - \upsilon_1' = \alpha \frac{1}{2} \sqrt{\frac{k_B T}{eU}}$$

from which the velocity compression factor

$$R = \frac{1}{2}\sqrt{\frac{k_B T}{eU}} \tag{5.6}$$

can directly be extracted. This can be also understood by the fact that the energy spread

$$\delta E = m \ \upsilon \ \delta \upsilon, \tag{5.7}$$

must stay constant during the acceleration. Consequently, if the velocity v increases, the spread  $\delta v$  has to decrease accordingly. Graphically this is illustrated in figure 5.1 in terms of  $\beta$ .

Since the Doppler broadening is proportional to the velocity distribution, the width must also be reduced by the factor R. Probing this accelerated ion beam along the z axis with a collimated, narrow linewidth laser, the width  $\Delta \omega_z$  of an absorption line, neglecting all other sources of line broadening as well as the natural linewidth, is

$$\Delta \omega_{\rm z} = \omega_0 \frac{v_1}{c} \cdot R \quad ,$$

where  $\omega_0$  is the non-shifted frequency of the absorption line in the rest frame [KAU76]. For a typical source temperature of 2000 K the linewidth for the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition



Figure 5.2: Standard setup for collinear spectroscopy on atoms [OTT89]. Ions are mass separated and sent into the COLLAPS beam line where they are superimposed with the collinear laser beam. In the charge exchange cell the ions are neutralized and the fast atoms enter the optical detection region, where the fluorescence light is detected.

of <sup>9</sup>Be becomes  $\Delta\omega_z \approx 5.2$  MHz at an acceleration voltage of 60 kV. Compared to the expected thermal linewidth  $\Delta\omega_z$  of 10 GHz [MAY00] in a thermal gas the advantage of this technique becomes obvious. Mostly, the theoretically predicted linewidth cannot be reached because the energy uncertainty in the source does not stem solely from thermal distributions. It is rather limited by the potential distribution inside the source than by the thermal energy distribution. Kaufmann's proposal was realized in many experimental setups in the last 30 years e.g., Oak Ridge [SPE81], Collaps [SCH78], Risiko [ZIM90] and is still attractive as recent proposals for collinear laser spectroscopy at the ANL [SAV08] or at the TRIGA reactor in Mainz or the Fair project indicate [KET08, NOE06].

As a result of this proposal the Collaps beam line at ISOLDE was set up in 1978/79 and will be described in detail in section 6.2. The typical setup of Collaps as used for spectroscopy on atoms is shown in figure 5.2 [OTT89]. The excitation energies of an ion of interest can be examined in two ways. Either by scanning the laser frequency at a fixed acceleration voltage, or, as in our case, by scanning the ion's acceleration voltage at a fixed laser frequency. In this case a signal from the fluorescence photons can be obtained in dependence of the ion's kinetic energy. The recorded spectrum has to be transformed into the frequency domain afterwards. At this point the main drawback of this technique appears: it is not possi-



Figure 5.1: The kinetic energy of an ion with mass  $m_0$  is plotted against its velocity  $\beta$ . The same distribution of the kinetic energy  $E_{kin}$  results in different velocity distributions  $\Delta\beta_1$  and  $\Delta\beta_2$  for slow and fast ions.

Isotope	U	$\beta$	$oldsymbol{ u}_{ ext{coll}}$	$\delta  u_{ m IS}^{ m art}$
7	60	$4.2849 \cdot 10^{-3}$	$0.9957 \cdot \nu_0$	
10	60	$3.5869 \cdot 10^{-3}$	$0.9964 \cdot \nu_0$	$-6.9257 \cdot 10^{-4} \cdot \nu_0$
7	59.994	$4.2847 \cdot 10^{-3}$	$0.9957 \cdot \nu_0$	
10	59.994	$3.5867 \cdot 10^{-3}$	$0.9964 \cdot \nu_0$	$-6.9254 \cdot 10^{-4} \cdot \nu_0$

Table 5.1: The isotope shift between <sup>7</sup>Be and <sup>10</sup>Be is calculated for 60 kV and for 59.994 kV. This small change in the acceleration voltage leads already to an uncertainty of approximately 8 MHz in the isotope shift determination of beryllium isotopes.

ble to extract the absolute excitation frequency for a transition because the acceleration voltage can not be determined with the required accuracy. For Be isotopes for example the kinetic energy of the ion has to be known within 1 V to determine the transition frequency with an accuracy of 30 to 40 MHz. The needed relative accuracy in the voltage measurement to determine it with an accuracy of approximately 1 MHz would be  $10^{-6}$  or  $10^{-5}$ .

For heavier isotopes, collinear laser spectroscopy can be used to determine the frequency shift between different isotopes or elements to a reference. For light isotopes, small fluctuations lead to tremendous velocity shifts and therewith strong shifts in the resonance frequency of the examined transition, as the following example illustrates on <sup>7</sup>Be and <sup>10</sup>Be: Assuming that there is no isotope shift between <sup>7</sup>Be and <sup>10</sup>Be one can derive  $\beta$  and the transition frequencies at 60 kV and at 59.994 kV to get an estimation of the artificially introduced isotope shift  $\delta \nu_{\rm IS}^{\rm art}$  as summarized in table 5.1. Subtraction of  $\delta \nu_{\rm IS}^{\rm art}$  for both acceleration voltages gives the uncertainty in isotope shift measurements

$$\left(-6.9257 \cdot 10^{-4} + 6.9254 \cdot 10^{-4}\right) \cdot \nu_0 = 3 \cdot 10^{-8} \cdot \nu_0, \tag{5.8}$$

which is only introduced by uncertainties of the acceleration voltage. Inserting a typical transition frequency  $\nu_0$  in equation 5.8 leads to an uncertainty of approximately 30 MHz for the isotope shift between <sup>7</sup>Be and <sup>10</sup>Be and still 8 MHz between <sup>10</sup>Be and <sup>11</sup>Be.

# 5.1.1 Overcoming the beam energy limitations of collinear laser spectroscopy

It was shown that the knowledge of the exact beam energy is crucial in collinear laser spectroscopy of light elements. Even if the accuracy of the ISOLDE high-voltage determination would be increased to the 1 ppm level - which is technically feasible these days [THU07] - there is still the limited knowledge of the exact starting potential of the laser ions inside the ion source. Hence, a high-precision beam energy measurement is required.

The idea of using laser spectroscopy in collinear geometry to determine ion beam energies, and thus to measure high voltages with high precision, was proposed by Poulsen [POU82] in the early eighties. This technique was suggested for the measurements of high voltages up to a few 100 kV to determine and calibrate the voltage of the electron cooler at



Figure 5.4: Frequency difference for parallel and anti-parallel isotope shift between isotope A and A'. Abbreviations are explained in the text.

the GSI experimental storage ring ESR. The capabilities and limitations of this aopproach were thoroughly investigated and discussed in [KNA00, GOE02]. The layout of the prototype allowed measurements up to 50 kV and the  $3d_{3/2,5/2} \rightarrow 4p_{3/2}$  transition in calcium ions was used for probing the ion velocity. It was demonstrated that accuracies in the order of  $10^{-4}$  can be reached and suggestions for further improvement were made. The theoretical lineshape of resonances in collinear laser spectroscopy was also discussed in this context [KRE04] and an improved wavemeter design for accurate laser frequency determination was proposed [KNA00]. The latter can in principle be replaced by compact femtosecond frequency combs as they are available nowadays [UDE02] provide even higher accuracies.

The lightest short-lived isotopes investigated by collinear laser spectroscopy to determine nuclear charge radii so far were the neon isotopes <sup>17</sup>Ne - <sup>28</sup>Ne [GEI00]. The method used to calibrate the energy in these measurements relied on an excitation scheme in V-configuration as shown in figure 5.3. Resonant excitation from a common ground state to two close lying excited levels was used to extract the beam energy in the following way: The Doppler shift in parallel and antiparallel configuration causes frequency shifts of opposite signs.

$$\nu_{\text{coll}} = \nu_0^{(1)} \gamma \left(1 + \beta\right) \qquad (5.9)$$

$$\nu_{\text{anti}} = \nu_0^{(2)} \gamma (1 - \beta) \qquad (5.10)$$

as shown in the figure. Hence, the kinetic energy eU



Figure 5.3: Excitation scheme used for the collinear /anti-collinear excitation in the beam-energy measurement. The dotted line represents the transition frequency in the rest frame. The colored arrows represent the Dopller shifted transition frequencies of the atoms [GEI02].

$$eU = \frac{mc^2 \left(\sqrt{\nu_{\text{coll}}} - \sqrt{\nu_{\text{anti}}}\right)^2}{2\sqrt{\nu_{\text{coll}} \cdot \nu_{\text{anti}}}}$$
(5.11)

can be found in which the two transitions occur at exactly the same laser frequency in

the laboratory system

$$\nu_{\rm coll} = \nu_{\rm anti} = \sqrt{\nu_{\rm coll} \cdot \nu_{\rm anti}}.$$
(5.12)

This technique requires two independent transitions which lie within the working range of the collinear spectroscopy setup. However, such a scheme cannot be applied in the case of the Be measurements since such transitions are not avilable in the Be<sup>+</sup> level scheme. Another alternative has been proposed by Borghs et al. [BOR81] also in the early eighties and was demonstrated in measurements on stable barium isotopes. The concept is shown in figure 5.4, where the following abbreviations are used

$$\nu_{\text{coll}} = \nu_0 \gamma \left( 1 + \beta \right) \tag{5.13}$$

$$\nu_{\text{anti}} = \nu_0 \gamma \left(1 - \beta\right) \tag{5.14}$$

$$D^A = \nu^A_{\text{coll}} - \nu^A_{\text{anti}} \tag{5.15}$$

$$\Delta_{\rm coll}^{AA'} = \nu_{\rm coll}^{A'} - \nu_{\rm coll}^{A} \tag{5.16}$$

$$\Delta_{\text{anti}}^{AA'} = \nu_{\text{anti}}^{A'} - \nu_{\text{anti}}^{A}$$
(5.17)

These equations can be modified and lead finally to an expression for the calculation of the isotope shift that is given by:

$$\delta\nu_{IS}^{AA'} \approx \left(\Delta_{\text{coll}}^{AA'} + \Delta_{\text{anti}}^{AA'}\right)/2 - Q^{AA'} \left(\Delta_{\text{coll}}^{AA'} - \Delta_{\text{anti}}^{AA'}\right)^2 \tag{5.18}$$

with

$$Q^{AA'} = \frac{1}{8\nu_0^A} \left( m_{A'}^{-0.5} + m_A^{-0.5} \right) / \left( m_{A'}^{-0.5} - m_A^{-0.5} \right).$$
(5.19)

In the demonstration measurements the collinear and anti-collinear measurements were carried out subsequently with a single laser system. It would of course be possible to perform similar measurements with two laser systems and rapid switching between them to observe the resonances quasi simultaneously as it is done in the approach described in this thesis. This would remove additional uncertainties due to possible voltage drifts between the measurements. However, the scheme proposed by Borghs et al. has never been applied on-line for short-lived isotopes. Our approach is based on absolute frequency measurements with a frequency comb and does not require the approximations made in the derivation of equation 5.18. Instead, the direct and relativistically correct relation

$$\nu_0^2 = \nu_{\rm coll} \cdot \nu_{\rm anti} \tag{5.20}$$

is used.

# Chapter 6 Laser spectroscopy at COLLAPS

The measurements of the nuclear charge radii of <sup>7,10,11</sup>Be were prepared in an off-line beam time in April 2008 and finally performed in an on-line run in June 2008 at the COLLAPS beam line at ISOLDE/ CERN. During the off-line beam time the experimental setup was tested and possible systematic error sources were investigated, whereas the final measurements of the radioactive isotopes were done in the on-line beam time. The general overview over collinear laser spectroscopy as given in 5.1 will be discussed in more detail in this chapter and the obtained results will be presented.

## 6.1 Ion source

A crucial point at nuclear facilities for the investigation of radioactive isotopes is the production process. ISOLDE at the European Research Center CERN uses the so-called *isotope separation on-line* technique. Here, high energy protons (1.4 GeV) are stopped in a thick target and radioactive isotopes are produced by nuclear reactions. The impinging protons cause spallation, fission and fragmentation processes, which lead to the production of the desired isotopes depending on the target material. For the production of beryllium isotopes the target material was uranium carbide.

Besides high production rates, a short transfer time of the produced isotopes from the target to the ionization region is important to allow experiments with short lived isotopes. Therefore, diffusion of radioactive isotopes out of the target, evaporation from the target surface, effusion to the ionization region and the ionization process itself need to be optimized. For beryllium, the uranium carbide target and the transfer line were heated to more than 2000°C. Behind the transfer line, the isotopes enter a second cylinder in which they are ionized by resonant laser excitation and ionization (RILIS). Because the ionization potential for Be is 9 eV, a surface ion source can not be used and the efficiency of the plasma ion source is not sufficient. A two step resonant ionization scheme into an auto ionizing state is used for beryllium. In table 6.1 the expected yields<sup>1</sup> for beryllium isotopes are listed.

A schematic of the RILIS setup at ISOLDE is shown in figure 6.1. The ionization takes place in a small area in front as well as in the cylinder which is put on high voltage

 $<sup>^{1}</sup>$  http://isolde.web.cern.ch/isolde



Figure 6.1: Illustration of ISOLDEs resonance ionization laser ion source RILIS. Further information can be found in [FED00] and the text.

of typically 30-60 kV. For efficient extraction of ions which have been produced in the cylinder, the applied voltage for heating (1.5 - 2 volts [SEB99]) is used as a guiding field - the positive potential is applied to the rear side of ionization tube as illustrated in figure 6.1. The ions are finally accelerated to ground potential, mass separated in the general purpose separator (GPS) with a mass resolution of  $\frac{M}{\Delta M} = 2400$  and sent into different experimental beam lines.

Mass	Half-life	Yield	Ionization
		ions/ $\mu {\rm C}$	
7	53 d	$1.4 \cdot 10^{10}$	RILIS
10	1.6 My	$6 \cdot 10^9$	RILIS
11	$13.8~\mathrm{s}$	$7 \cdot 10^6$	RILIS
12	$21.3 \mathrm{\ ms}$	$1.5 \cdot 10^3$	RILIS
14	4.4 ms	4	RILIS

Table 6.1: Specified yields for Be isotopes for 1.4 GeV protons impinging on a uranium carbide target and ionization in the RILIS ion source.



Figure 6.2: Illustration of the COLLAPS beamline including the applied voltages. Further details can be found in the text.

## 6.2 COLLAPS beam line

The COLLAPS beam line was set up in 1978/79 at ISOLDE. The beam line for collinear laser spectroscopy, as it is briefly described in chapter 5.1, was optimized to fulfill the requirements for collinear/ anti-collinear precision spectroscopy. Those elements that are of importance for the beryllium spectroscopy are depicted in more detail in figure 6.2: As normal, the ion beam enters the beam line through a 10° bender that is needed to superimpose it with the collinear laser beam. It passes electro static deflectors for ion beam steering and a quadrupole triplet for beam focussing. Since spectroscopy is performed on beryllium ions, a charge exchange cell is not required and it was therefore replaced by a set of apertures which are designed to reduce stray light in the optical detection region as indicated in figure 6.2. Behind the region for optical detection of fluorescence light the ion beam passes another set of apertures, which is used for stray light reduction of the anti-collinear laser beam. Directly behind the optical detection region a Faraday cup can be moved into the beam line to detect and measure ion beam currents down to  $10^{-12}$  A. Two variable irises are inserted into the beam line with a distance of approximately 3 m to ensure that ion and laser beams are superimposed and parallel.

Floating Fluke high voltage power supplies are used to apply a fixed voltage up to  $\pm 10$  kV to the optical detection region. At its entrance, a set of deceleration electrodes is placed for smooth acceleration or deceleration of the ion beam. Voltages for the different electrodes are created by dividing the potential of the optical detection region with a fixed ratio voltage divider. To perform fast Doppler tuning across the resonance pattern, the Fluke HV supplies can be floated to a potential of  $\pm 500$  V. This voltage is controlled by the data acquisition computer. An 18 bit digital to analog (DAC) converter board generates a DC voltage between  $\pm 10$  V, which is fed into a Kepco high voltage amplifier and amplified by a factor of approximately 50. The amplification factor has to be measured several times during the beam time to ensure exact knowledge of the beam energy. An additional offset voltage of typically 200 V is applied to the deceleration/ acceleration electrodes. It is is



Figure 6.3: Two dye lasers operated at 624 nm and 628 nm, respectively. The first one is locked to hyperfine transitions of  $I_2$  molecules (left) whereas the second is directly locked to a frequency comb (right). Both outputs are frequency-doubled to reach the frequencies in the ultraviloett region required for the Be<sup>+</sup> transition.

used to reduce optical pumping into dark states and recoil effects in front of the optical detection region. The applied voltage at the detection region is measurable with a high voltage divider (Type: Julie Research) that has a dividing ratio of 1:1000 and a digital multimeter (Type: Prema) with a precision of about  $10^{-4}$ . The result of the high voltage measurement is read out by the data acquisition system.

## 6.3 Laser system

The laser system used for spectroscopy consists of two dye lasers, a frequency comb and two frequency doublers. An overview of this system is shown in figure 6.3, its setup, development and performance is described in detail in [ZAK09] and [KRI08, NOT07] and will be covered here only briefly. The two dye lasers were pumped with 8 Watt at 532 nm provided by Coherent Nd:YAG lasers (Verdi V8, V18). The dye lasers (Coherent 699) were operated with Sulfuro-Rhodamin and had an output power of approximately 500 mW at 626 nm. One of the dye lasers was locked to a fiber frequency comb from Menlo Systems, whereas the second one was locked to various hyperfine transitions of the I<sub>2</sub> molecule. The light of the dye lasers was transported to two frequency doublers near the COLLAPS beam line using photonic crystal fibers, which provided a transmission efficiency of 40 to 50 %. The generated second harmonic light passed some beam shaping optics to achieve a beam focus of approximately 3 mm in the optical detection region and was sent into the beam line in collinear and anti-collinear direction.

## 6.3.1 Locking to a frequency comb

Frequency offset locking is a common tool for laser stabilization. The beat frequency between a reference laser with a known frequency and a second laser is used to produce the stabilization signal. A frequency comb can be understood as a few thousand reference lasers, with exactly known frequencies. As the technique of frequency offset locking as



Figure 6.4: (a) A travelling femto second laser puls in a resonator, which emits pulses with the repetition frequency  $\nu_r = \tau^{-1}$ . (b) Fourier transformation of the electrical field, which consits of thousands of narrow frequencies, which are spaced by by  $\omega_R = 2\pi\nu_r$  - the so-called frequency comb. The carrier envelope offset  $\omega_{CE}$  is the offset of the first mode from the y axis.

well as the frequency comb technique are described in detail in literature [SCH99, HOL00] only the experimental setup and the frequency comb will be described in this section.

The frequency comb which was used for the long time stabilization of the anti-collinear dye laser and also for the determination of the absolute transition frequencies of the iodine lines used for spectroscopy is based on a femto second fiber laser. Figure 6.4 (a) illustrates a traveling femto-second laser puls train. The Fourier transformation of such a femto second puls is a spectrum of equally spaced modes, which are shown in figure 6.4 (b). The distance between two modes is given by the repetition rate  $\omega_{\rm R}$  of the fiber laser. The propagation velocity of the envelope of the electric field and that of the phase of the electric field is slightly different which leads to a phase slip from puls to puls, indicated as  $\Delta \varphi$  in figure 6.4 (a). In the frequency domain, this phase slip or carrier envelope offset is represented by an offset  $\omega_{\rm CE}$  from zero when the comb modes are traced back towards the y axis. The basis technique of the frequency comb is to fix and stabilize the repetition rate  $\omega_{\rm R}$  and this offset  $\omega_{\rm CE}$ . Therewith it is possible to calculate the frequency of any given comb mode  $\omega_n$  using the relation

$$\omega_{\rm n} = n \cdot \omega_{\rm R} \pm \omega_{\rm CE} \tag{6.1}$$

In the setup used here the frequency comb beam is combined with a small part of the output of the dye laser that has to be stabilized to the frequency comb. A grating is used to remove the spectral part of the comb that is not in the dye laser region and the two laser beams are superimposed on a fast photo diode. The beat frequency with the closest comb mode is measured and stabilized using phase locking of the beat signal. The comb mode n is determined with a high resolution wavemeter (Type: HighFinesse). The frequency of the dye laser can then be calculated according to

$$\omega = n \cdot \omega_{\rm R} \pm \omega_{\rm CE} \pm \omega_{\rm beat} \tag{6.2}$$



Figure 6.5: Schematics of the FM saturation lock to hyperfine transitions in iodine molecules. The dye laser beam is devided into a probe and a pump beam. The probe beam is frequency modulated in an EOM, while the probe beam is phase shifted and amplitude modulated in an AOM. The probe beam is detected after transmission through the iodine cell and demodulated at the EOM frequency. A Lock-in amplifier is used to increase the signal to noise ratio by phase-sensitive detection at the AOM amplitude modulation frequency.

### 6.3.2 Locking to $I_2$ hyperfine transitions

Iodine molecules have wide rotational and vibrational bands, which lead to a complex hyperfine structure with transition frequencies in the VIS and IR region. Many of the resulting electronic transitions have been measured and can be calculated with relatively high accuracy. They are widely used for laser stabilization. In this section an overview about the direct stabilization of a dye laser to iodine transitions by *frequency modulation saturation spectroscopy* (FMS) will be given. This is a well described technique and only the used setup will be described. More detailed information to this subject can be found for example in [GEH85, KRI08].

To resolve the hyperfine structure of iodine molecules in a gas cell by laser spectroscopy the Doppler broadening has to be eliminated. A strongly simplified sketch of the setup for FMS is illustrated and explained in figure 6.5. In case of an interaction between pump and probe beam with the same molecular velocity class, the depopulation of the involved energy levels causes an alteration of the absorption and dispersion signal of the probe beam. These changes can be used by phase sensitive analysis of the probe beam for stabilization.

The dye laser beam is divided into a probe and a pump beam that have orthogonal polarization direction. The probe beam is sent into an electro-optical modulator (EOM) where two sidebands at  $\omega_{\text{Dye}} \pm \Omega_{\text{EOM}}$  are generated. Since the EOM produces pure frequency modulation, the sidebands are balanced. They have equal amplitudes and opposite sign, and no intensity modulation is observed on a fast photo diode. The pump beam passes through an accusto-optical modulator (AOM) where it is frequency shifted by a fixed frequency  $\Omega_{\text{AOM}}$  and amplitude modulated with a frequency  $\Omega_{\text{AM}}$ . Pump and probe beam are superimposed in the iodine gas cell and separated again by their polarization direction. The probe beam is sent to a fast photo diode, whereas the pump beam is dumped.

The principle of (FMS) is the following: As long as pump and probe beam do not interact with the same molecules and with different velocity classes, the two sidebands of the probe beam will only detect the slow varying Doppler broadened transition that covers the whole hyperfine structure. In this case there is practically no difference between absorption and dispersion of the two sidebands. However, close to resonance of a hyperfine transition the pump laser saturates the transition of one velocity class and if one of the sidebands of the probe beam interacts with the same molecule it is less scattered than the other sideband. This removes the balance of the sideband and a beat signal between the sidebands and the carrier will be observed at the frequency  $\Omega_{\text{EOM}}$ . The signal is demodulated and a dispersion like signal can be obtained by choosing the right phase shift  $\Delta \varphi_{\rm EOM}$  for the local oscillator input of the mixer (compare with figure 6.5) The demodulated signal is fed into a Lock-in amplifier to obtain a better signal to noise ratio of the spectroscopy signal. All iodine lines that were used in the experiment started from a rotational level with even J. Those transitions exhibit a hyperfine structure splitting with 15 components [KRI08] as illustrated in figure 6.5 (b). The  $a_1$  component is in all cases separated and simplifies the identification and reliable locking during the beam time.

All Iodine lines, which have been used during the experiments at ISOLDE, have been measured several times before, during and after the beam times and their absolute tran-

No.	HFS	Frequency	
		MHz	
1	R(62)(8-3)a1	479 804 354.9 (4)	
2	R(70)(10-4)a1	$479 \ 823 \ 072.3 \ (4)$	
3	P(64)(10-4)a1	479 835 708.7 (4)	
4	R(60)(8-3)a1	$479\ 870\ 012.0\ (4)$	
5	R(58)(8-3)a1	$479 \ 933 \ 416.0 \ (4)$	
7	R(54)(8-3)a1	$480\ 053\ 468.8\ (4)$	
8	R(52)(8-3)a1	$480\ 110\ 119.4\ (4)$	
9	R(50)(8-3)a1	$480\ 266\ 578.4\ (4)$	
10	R(48)(8-3)a1	$480 \ 314 \ 236.6 \ (4)$	
11	R(42)(8-3)a1	$480 \ 359 \ 648.9 \ (4)$	
12	R(40)(8-3)a1	$480 \ 402 \ 815.6 \ (4)$	
13	R(38)(8-3)a1	$480 \ 443 \ 737.9 \ (4)$	
<b>14</b>	R(36)(8-3)a1	$480 \ 482 \ 416.3 \ (4)$	

Table 6.2: Measured transition frequencies of the iodine hyperfine transitions used in the COLLAPS measurements [KRI08].

sition frequencies were determined with the frequency comb with an uncertainty, that includes statistical and systematic error sources, of approximately 400 kHz and are summarized in table 6.2 [KRI08].

## 6.3.3 Frequency doubling

Frequency doubling is a non linear optical process, which occurs in special crystals and is widely used in laser physics. The efficiency  $E_{\rm SHG}$  of such processes as in BBO<sup>2</sup> and LBO<sup>3</sup> crystals is strongly dependent from the incident laser power at the fundamental wave length. It can be derived with

$$E_{\rm SHG} = \frac{P_{2\omega}}{P_{\omega}} = l^2 \cdot K \cdot \frac{P_{\omega}}{A}, \tag{6.3}$$

where P is the power of the second harmonic and the fundamental wavelength, l is the crystal length, K is a material dependent factor and A is the focal size of the fundamental laser beam. As the single path doubling efficiency would be only 0.01 % for cw-lasers, the power has to be enhanced in a cavity. Two different frequency doublers, one with a bow-tie resonator from Tekhnoscan and the other with a delta resonator from SpectraPhysics, were used in the experiment. Both frequency doublers are illustrated in figure 6.6.

 $<sup>^{2}</sup>$ Barium borate

<sup>&</sup>lt;sup>3</sup>Lithium triborate



Figure 6.6: Schematic view of (a) FD-SD-07 from Tekhnoscan, (b) Wavetrain from SpectraPhysics.

Figure 6.6.(a) shows the frequency doubler FD-SF-07 from Tekhnoscan. The fundamental passes a lens for *mode matching* to the cavity mode and is directed with two mirrors into the cavity. It consists of four mirrors, which guide the fundamental light in a closed loop through the crystal. Mirror M4 is the output window for the second harmonic (and residual light of the fundamental), whereas mirrors M2 and M3 are mounted on piezos for the compensation of slow and fast frequency fluctuations by changing the cavity length. The stabilization signal is generated with the Hänsch-Couilluad technique, which requires non-degenerated polarization modes as typically obtained by using a cavity with birefringent materials [MAN]. Therefore light of "wrong" polarization cannot enter the cavity and is directly reflected by mirror M1. A part of the fundamental light in the resonator leaves the cavity through the entrance window after one round trip. The phase shift gathered during the round trip can be analyzed by comparing it to the phase from the directly reflected light which serves as a phase reference. Therefore the light is analyzed with two photodiodes and polarizing beam splitters outside the cavity. This technique allows a wide scan range of the fundamental over several GHz. More information about the used frequency doubler and the Hänsch-Couillaud stabilization can be found in the user manual and [HAE80].

Figure 6.6.(b) shows the frequency doubler Wavetrain from SpectraPhysics. After the mode matching lens the fundamental passes a  $\lambda/2$  plate and an EOM. The EOM is operated at 80 MHz and is used to generate sidebands on the fundamental. Afterwards the fundamental is guided into the resonator, which consists of only two mirrors and a prism on a piezo mount, which is used for the stabilization of the cavity length. The error signal is generated with the Pound-Drever-Hall technique [DRE83]. It analyses the interference pattern of the fundamental laser beam, which is partially reflected at the entrance window and the leak light, that leaves the cavity in resonance to create the stabilization signal. This technique allows wide scans of the fundamental over some GHz.



Figure 6.7: Coupling of the laser system as shown in figure 6.3 to the COLLAPS beam line: The laser beams are fiber coupled and transported close to the beam line, where frequency-doublers were used to create uv light at 314 nm and 312 nm, respectively. Both lasers were directed into the beam line and were superimposed with each other and the ion beam. Shutters can be used to block the laser beams.

## 6.4 Measurement scheme

A schematic of the whole experimental setup at COLLAPS is shown in figure 6.7. Both frequency doubled dye laser beams are directed into the COLLAPS beam line. Even though it is in principle possible to record fluorescence in collinear and anti-collinear direction at the same time it is more convenient to record them separately. For this purpose fast shutters were used which blocked the laser beams in turns. The signal of the data acquisition system that is used for switching the shutters is called *outbit* and has a high and a low level of 5 V or 0 V, respectively and is toggled whenever a scan over the hyperfine structure is completed. The outbit and its inverted signal as well as the photomultiplier signals are connected to coincidence units, where the recorded spectra for collinear and anti-collinear di-



Figure 6.8: The data acquisition computer toggles the outbit whenever a scan over the hyperfine structure is finished. Depending on the outbit, the photomultiplier signals are sorted into scaler 1 and 2 or scaler 3 and 4, respectively. The whole procedure is repeated serveral times until sufficient statistics for the spectrum is obtained.

rection are sorted as illustrated in figure 6.8. As already mentioned, it is crucial to record the spectra for both directions at the same acceleration voltage. This can be assured by summing approximately 50 spectra for isotopes with high yields, and 200 or more for those with low yields. With this technique fast fluctuations of the acceleration voltage are averaged, while slow voltage drifts would result in line broadening.

Using  ${}^{9}\text{Be}^{+}$  as an example the following section will describe how the right acceleration voltage, scan region and laser frequencies have to be chosen. The operation voltage of ISOLDE is usually set to 50 or 60 kV. The Fluke power supplies can be set to  $\pm 10$  kV at maximum and the scanning range is  $\pm 500$  V. Assuming that the operation voltage is 60 kV this means that the ion's kinetic energy can be chosen between 49.5 and 70.5 keV. According to

$$E_{\rm kin} = (\gamma - 1) \cdot m_o c^2 \tag{6.4}$$

this corresponds according to the Doppler shift

$$\nu_{\rm coll} = \nu_0 \gamma (1 + \beta) \tag{6.5}$$

to a frequency range from 960492.418 GHz to 961130.597 GHz for the collinear laser frequency. All iodine lines within this frequency range of the fundamental can be used for locking, but to avoid operating at the maximum output power of the high-voltage devices iodine lines at the edge of this frequency range are usually avoided. Comparing the frequency range with table 6.2 shows that iodine lines 8 to 13 could be used for locking at an acceleration voltage of 60 kV. Once an iodine line is chosen, having in background that this frequency defines the frequency of the collinear laser, the needed voltage U can be calculated by modifying equation 6.5 to

$$\beta = \frac{\left(\frac{\nu_{\text{iodine}}}{\nu_0}\right)^2 - 1}{1 + \left(\frac{\nu_{\text{iodine}}}{\nu_0}\right)^2} \tag{6.6}$$

and inserting it into equation 6.4. With the now known acceleration voltage it is possible to calculate a scaling factor  $k_{\text{scaling}}$ . Assuming that the conversion between scanning voltage and frequency shift is linear over the whole hyperfine structure (the error is in the order of a few kHz) it is possible to translate the voltage scan directly into a frequency scan. It is given by

$$k_{\text{scaling}} = \gamma \beta \nu_0 \left( 1 - \sqrt{1 - \frac{\Delta U}{U}} \right) \tag{6.7}$$

with units in  $\frac{MHz}{\Delta U}$ . Translating the lowest and highest transitions of the hyperfine structure into voltages and dividing it through the Kepco amplification factor defines the scan region for the 18 bit DAC. The anti-collinear laser, is locked to a frequency, which is calculated by

$$\nu_{\text{anti}} = \nu_0 \gamma (1 - \beta) + \nu_{\text{HFS}}^{\text{lowest}}.$$
(6.8)

The value for  $\beta$  and  $\gamma$  has been calculated before and leads directly to the locking frequency of the anti-collinear laser. With these settings a voltage scan will shift the velocity and therewith the resonance frequency of the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition of <sup>9</sup>Be<sup>+</sup> to higher frequencies for the collinear laser and to lower frequencies for the anti-collinear laser.



Figure 6.9: The obtained spectra of  ${}^{9}\text{Be}^{+}$  at 30 and 60 kV ISOLDE voltage clearly show side peaks on the left and right side of the hyperfine transitions. Peaks on the left appear due to coulomb interaction of the ion beam with residual gas, while those on the right may arrise from ion source settings. At 60 kV there is still a hint of the peak on the right side but it is not as prominent as at 30 kV. More discussion in the text.

Figure 6.9 shows typical spectra that were obtained at an acceleration voltage of 60 kV and 30 kV. The x-axis gives the Doppler tuning voltage as it has been recorded by the data acquisition system. Already at 60 kV it is clearly observable, that besides the 4 peaks of the hyperfine structure additional structures arise. This is much more pronounced in the spectra at 30 kV, which were taken for a closer inspection of these effects and were only analyzed to determine the fitting function and will be described in the following section.

## 6.5 Data analysis

To understand the fit function which has been chosen for the data analysis spectra for  ${}^{9}\text{Be}^{+}$  recorded at an acceleration voltage of 30 kV have to be discussed. They are the worst spectra that were recorded, but nevertheless helpful to understand the peak structure of the obtained spectra during off- and on-line beam time. Measurements at 30 kV have only been used for the preparation of the fit function but were not considered in further

analysis.

Examples for collinear and anti-collinear spectra are shown in figure 6.9. All expected hyperfine transitions for  ${}^{9}\text{Be}^{+}$  are clearly resolved, but both spectra show satellite peaks on the left and right side of each hyperfine transition. A first fitting with nine individual Voigt profiles gave evidence that these satellite peaks have always the same distance to the corresponding main peak. The left satellite peak is separated by -4 volts from the hyperfine transition, while the right satellite peak has a distance of 1.5 to 2 volts to the corresponding main peak. As these peaks appear on the same side in the collinear and anti-collinear spectrum they represent a difference in the kinetic energy of the ions. The peak on the left side is caused by ions which are slower, whereas peaks on the right side are caused by ions which are faster than the majority of ions.

The peak on the left side is an often observed phenomenon in collinear spectroscopy. Due to inelastic collisions of fast ions with residual gas atoms the Be ions can get excited. The energy required for this process is taken from the ion's kinetic energy and afterwards radiated in form of photons. The lowest possible excitation in Be<sup>+</sup> ions os the  $2s \rightarrow 2p$ transition, which has an energy of 4 eV, is in excellent agreement with the peak distance. The origin of the right satellite peak is not fully understood. Ions with a higher energy could be created at a different potential in the ion source. It is possible, that ions which have been created inside the ionization tube have a higher potential energy due to the heating voltage which is used as guiding field as mentioned in section 6.1.

Fitting is performed using the package Minuit, which is embedded in the program MCP that controls the data acquisition at the COLLAPS experiment. At the start of the fitting routine the number of hyperfine transitions is calculated based on the nuclear spin and the angular momentum of the electron state. The peak positions of the hyperfine components are not fitted independently and the Casimir formula 3.1 is used to calculate the positions of the hyperfine peak according to the interval factors  $A_{2s}$  and  $A_{2p}$ . The following parameters are left free during fitting:  $A_{2s}$  and  $A_{2p}$ , the Gaussian linewidth, the intensity of the four main peaks and the center of gravity for each isotope. Constraints were used for the following parameters: The Lorentzian linewidth is fixed to the natural linewidth of 19.64 MHz, the distance between main and side peaks is -4 and +2 V, respectively and the amplitude ratio between side and main peaks is the same for all hyperfine transitions. All hyperfine transitions exhibit a Voigt profile with two side peaks as described above. For best accuracy the fit takes place in the frequency domain, where the frequency axis is calculated based on the relativistic expressions and not on the linear approximation with  $k_{\text{scaling}}$ .

From figure 6.9 it is obvious, that the center of gravity obtained in the fit does not coincide for the collinear and anti-collinear spectrum. The difference occurs due to the lock frequency of the anti-collinear laser. It would be only possible to have the two centers at exactly the same point, if all frequencies are known beforehand. Since this is not the case, but the formula

$$\nu_0^2 = \nu_{\text{anti}} \cdot \nu_{\text{coll}} \tag{6.9}$$

requires a measurement at the same ion's kinetic energy, a method must be found to correct the difference in the analysis. The distance

$$\Delta U = U_{\rm coll}^{\rm cg} - U_{\rm anti}^{\rm cg} \tag{6.10}$$

between the center of gravity in collinear and anti-collinear spectra can be directly converted into a frequency difference  $\Delta \nu$  by

$$\Delta \nu = \Delta U \cdot k_{\text{scaling}}.$$
(6.11)

Instead of tuning the anti-collinear laser frequency it is more convenient to artificially shift the laser frequency afterwards. Now equation 5.20 can be modified to:

$$\nu_0 = \sqrt{\nu_{\text{anti}} \cdot (\nu_{\text{coll}} - \Delta \nu)}.$$
(6.12)

It should be noted, that the knowledge of the total acceleration voltage U is required at this point of the analysis. But the uncertainty introduced with a relative uncertainty of  $10^{-4}$  is in the order of 100 kHz and therefore negligible since the difference between the center of gravities in collinear and anti-collinear spectra is usually smaller than 5 V.

## 6.6 Results of the off-line beam time

During the off-line beam time the experimental setup and the scanning and data acquisition procedures were systematically investigated to identify and estimate possible sources of systematic frequency shifts. In total, 88 spectra for <sup>9</sup>Be<sup>+</sup> at 60 kV were analyzed and will be described in this paragraph. For each measurement set, the individual results are compared to the overall average of all 88 spectra, which is  $\nu_0^{all} = 957199552.6(5)$  MHz. Also the statistical variation over all 88 measurements represented by the standard deviation of 0.97 MHz is shown for comparison.

#### 6.6.1 Investigation of systematic effects

Errors as discussed in this section are statistical and given by the standard deviation if more than one data set was available. In cases, where only single data points are analyzed the error is conservatively estimated to be the standard deviation of all analyzed measurements of 0.97 MHz as given above. One should remember that the transversal ion-beam profile along the beam line could not be analyzed. Only the transmittance through the inserted apertures can be used to estimate that the ion beam propagates almost parallel along the the symmetry axis of the beam line with a diameter of less than 7 mm, which is given by aperture for stray light reduction.

#### 6.6.1.1 Different iodine lines

For a fixed ISOLDE voltage, as it was always used in these measurements, the different iodine lines required different acceleration and deceleration voltages at the detection region according to section. 6.4. Wether this voltage change has an influence on the ion beam that causes a shift of the measured transition frequency the absolute transition frequencies were determined for all available iodine lines. The results are shown in figure 6.10 (a). For each iodine line the mean value of the analyzed data, between 3 and 73 individual measurements could be analyzed, is shown with an uncertainty given by the obtained standard deviation. Although a voltage range of almost 10 kV is covered, systematic



Figure 6.10: Dependence of the absolute transition frequency of the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition on various parameters. The thick line represents the average over all 88 analyzed measurements and the dashed lines the standard deviation of these measurements. (a) Lock of the collinear laser to different iodine lines. (b) Different powers of the anti-collinear laser and (c) Different laser beam diameters of the collinear laser. Further discussion in the text.

effects do not appear. For iodine line 13 the literature value was used instead of the value that is listed in table 6.2, because an inconsistency was found during data analysis that is still under evaluation.

#### 6.6.1.2 Power dependence and laser focus

For the determination of the influence of the laser power and the laser beam profile on the resonance profile and the central frequency the laser power of the anti-collinear laser was varied in a wide range from 0.6 mW to 5 mW, while the power of the collinear laser was approximately 0.6 mW in all measurements. A higher intensity can affect the lineshape in two ways: It can cause power broadening - which is less severe for our purposes - but it can also shift the resonance center since an increasing number of scattering events changes the ion velocity and therewith the resonance frequency. The results of this series of measurements are shown in figure 6.10 (b). Also here a systematic effect is not observed. Evaluation of the resonance frequency was performed by fitting the line profile as described in the previous chapter. Since the Lorentzian lineshape was kept fixed, the fitting routine increased the Doppler width. The reduced  $\chi^2$  did not change significantly in the measurement series, indicating that the line profile can still be reasonably well reproduced.

A second series of experiments was used to find the most appropriate focal size of the collinear laser beam at the optical detection region. Here, both lasers had a power of approximately 1 mW and the focus of the collinear laser was set to 1, 3 and 7 mm, respectively, in the optical detection region. Again a systematic effect on the absolute transition frequency could not be observed as shown in figure 6.10 (c). The best signal to noise ratio was found for a beam diameter of 3 mm which was used in further measurements.

#### 6.6.1.3 Ion beam focusing

Also the focus of the ion beam was changed to find an optimal signal to noise ratio and to investigate a possible influence on the observed transition frequencies. 6 measurements were performed for 2 different quadrupole settings. Combined result of these measurements is a value of  $\nu_0 = 957199552.3(2)$  MHz, which is in excellent agreement with the overall result of  $\nu_0^{all} = 957199552.6(5)$  MHz.

#### 6.6.1.4 Offset voltage

The offset voltage between the deceleration electrodes and the detection region has been varied in a range of  $\pm 200$  V to test the influence of this potential on the resonance frequencies. The main effect that can be expected is that the ions start to interact with the laser already before they enter the detection region. This would have two consequences: The ions would suffer recoil from the absorption and emission process and, hence, the ion velocity would have changed when they enter the detection region. This would result in a symmetric shift to higher frequencies for collinear and for anti-collinear spectra. Such a shift is particularly dangerous, because it does not cancel when the rest-frame frequencies are calculated. The second effect that would occur in such a case is a reduction of the



Figure 6.11: Dependence of the absolute transition frequency of the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition on various parameters. The thick line represents the average over all 88 anlyzed measurements and the dashed lines the standard deviation of these measurements. (a) Variation of the offset voltage. (b) Misallignment of the collinear laser relative to the anticollinear laser in horizontal and vertical direction. On the right the reference measurements are shown. (c) Misallignment of the ion beam in vertical and horizontal direction relative to the laser beam axis. Further discussion in the text.



Figure 6.12: Different kinds of laser-laser and laser-ion beam alignment. (a) One of the lasers is perfectly overlapped with the ion beam, while the second one crosses them under a small angle. (b) Both laser are perfectly overlapped, while the ion beam crosses the laser beams under a small angle.

signal intensity due to the depopulation of the probed hyperfine state in odd isotopes as described in section 3.1. These effects should appear particularly strong at an offset voltage of 0 V, because in this case the ions can get into resonance with the laser frequency approximately 20-30 cm in front of the optical detection region. The intensity ratio of the different transitions should change in such a way that the weaker transitions appear more prominent than expected.

Figure 6.11 (a) shows the obtained results. All three points are above the global average, but within the uncertainty and absolutely no variation of the transition frequency is observed. The peak intensities in the spectra show fluctuations that cannot be correlated with the offset voltage and are probably a result of beam intensity fluctuations. The result indicates that the ion beam is quite diffuse and that the laser beams interact only a few times with the same ion

#### 6.6.1.5 Misalignment

A misalignment of one or both laser beams relative to the ion beam, as illustrated in figure 6.12, will lead to large shifts of the center frequencies and is expected to be one of the main contributions to the total systematic uncertainty. The anti-collinear laser in figure 6.12 (a) is perfectly overlapped with the ion beam, whereas the collinear laser enters under a small angle. The absolute transition frequency has to be derived by

$$\nu_{\text{coll}} = \nu_0 \gamma (1 + \beta \cos \alpha) \tag{6.13}$$

$$\approx \nu_0 \gamma (1 + \beta (1 - \alpha^2)) \tag{6.14}$$

$$\approx \nu_0 \gamma (1+\beta) - \nu_0 \gamma \beta \alpha^2 \tag{6.15}$$

This introduces for  $\alpha = 1$  mrad and  $\beta = 0.004$  a shift of the derived rest frame frequency of about :

$$\frac{1}{2}\nu_0\gamma\beta\alpha^2 \approx \frac{1}{2}\nu_0\beta\alpha^2 \approx \frac{1}{2}\cdot 4\cdot 10^{-3}\cdot 10^{-6}\nu_0 \approx 2 \text{ MHz}, \tag{6.16}$$

which is quite severe.

In the second case - shown in figure 6.12 (b) - both lasers are very well aligned but a misalignment between the ion beam and the two laser beams leads to a shift of the collinear resonance to lower frequencies, while the anti-collinear laser is shifted to higher frequencies according to  $\nu_{\text{anti,coll}} = \nu_0 \gamma (1 \pm \beta \cos \alpha)$ . In first order, the two shifts cancel out while in an exact treatment the the shift is on the order of a few kHz under the same conditions as above. Hence a very good overlap between the two laser beams is much more important, than the exact overlap between the two laser beams and the ion beam. Fortunately, the laser beam overlap can be ensured over a much longer distance than the overlap with the ion beam and therefore a value considerably below 1 mrad should be obtainable. To guarantee such a good overlap of laser and ion beam during the experiments two adjustable irises were inserted in the beam line.

For the test of case (a) both irises were opened and the collinear laser was misaligned by approximately  $\pm 3$  mm over a distance of 6 m in vertical and horizontal direction. This corresponds to an angle of approximately 0.5 mrad. All measurements have been repeated at least twice and between consecutive misalignments resonances with re-optimized laser beam overlap were recorded. The results of this study are shown in figure 6.11 (b). They show that the misalignment is shifting the obtained transition frequency in the scale of  $\pm 0.5$  to  $\pm 1$  MHz. Theoretically the obtained frequency shift should be the same for a horizontal and vertical misalignment. Even though this is not completely understood, the laser beam alignment seems to have some effect on the absolute transition frequency and a systematic uncertainty in the order of 500 kHz is estimated.

For the test of case (b), both lasers were overlapped as good as possible and the ion beam was steered in vertical and horizontal direction by adjusting the potentials of the ion optic elements at the beginning of the beam line. Since further beam diagnostics were not available it is hard to estimate the degree of tilting of the ion beam. However, the observed effect, as shown in figure 6.11 (c), is again quite small. In vertical direction all points coincide with the global average, whereas there are some deviations for the horizontal misalignment. While these measurements for the horizontal misalignment were taken the beam intensity was fluctuating between 500 pA to 1.2 nA and a realignment of the RILIS ion source was necessary. This can probably result in another beam profile and beam path that could explain the observed deviations. The misalignment uncertainty of 500 kHz as mentioned above should be large enough to cover both contributions, so that no additional uncertainty is introduced.

#### 6.6.2 Summary of uncertainties

In this section the final uncertainties as used for data analysis of the on-line beam time will be discussed. One has to distinguish between the error for the absolute transition frequencies, the error of the isotope shift determination and the error for the determination of A factors. For the absolute transition frequency three main parts have to be taken into account:

1. Systematic uncertainties which arise from the experimental setup as analyzed above. The only systematic effect was observed for laser-ion beam misalignments and is estimated to:

$$\Delta \nu_{\rm align} = 500 \text{ kHz} \tag{6.17}$$

2. The frequency comb respectively the frequency reference of the frequency comb. For the stabilization of the repetition rate a frequency reference is needed. During the experimental run a Rb clock (Stanford Research Systems FS725) with a specified uncertainty of  $\pm 5 \cdot 10^{-11}$  and an additional uncertainty due to aging of  $< 5 \cdot 10^{-10} \frac{1}{a}$  has been used. As described in [KRI08] the overall uncertainty for the determination of the absolute transition frequency of the iodine lines as well as for the direct lock to the frequency comb is approximately 410 kHz, which results in an uncertainty of

$$\Delta \nu_{\rm fc} = 820 \text{ kHz} \tag{6.18}$$

in the second harmonic.

3. Recoil shifts appear due to repeated absorption of photons, which will accelerate (collinear laser) or decelerate (anti-collinear laser) Be<sup>+</sup> ions. In both cases the resonance is shifted to higher laser frequencies and the geometrical average will be shifted accordingly. For odd isotopes this effect is smaller than for even ones, because after few interactions dark states will be populated as mentioned in section 3.1. Absorption of a single photon induces a change of the resonance frequency for <sup>7,9,10,11</sup>Be<sup>+</sup> of about 600, 470, 420 and 383 kHz, respectively. For a diffuse ion beam that has only a short interaction time with the laser field it is reasonable to estimate that odd isotopes absorb in average 1.5 and <sup>10</sup>Be<sup>+</sup> 3 photons. This assumption is fortified by the fact, that no allusion to recoil effects, which are strongly power dependent, could be observed during the off- and on-line beam time. The estimated error is

$$\Delta \nu_{\rm rec} = 1.26 \text{ MHz} \tag{6.19}$$

All three uncertainties are systematic, however, they are uncorrelated concerning the direction of the shift. Hence, they can be combined with the statistical uncertainty by taking the geometrical average. In summary the total uncertainty of the absolute transition frequency of isotope A can be calculated by:

$$\Delta \nu_0^A = \sqrt{\left(\text{sem}(\nu_0)\right)^2 + \left(\Delta \nu_{\text{align}}\right)^2 + \left(\Delta \nu_{\text{fc}}\right)^2 + \left(\Delta \nu_{\text{rc}}\right)^2}$$
(6.20)

with the standard error of the mean sem( $\nu_0$ ). In the calculation of the uncertainty of the isotope shift  $\delta\nu_{\rm IS}^{\rm AA'}$  between isotope A and A' frequency comb related uncertainties cancel out and the error based on recoil effects has to be treated differentially and becomes therewith smaller.  $\Delta\nu_{\rm rc}^{\rm AA'}$  is approximately 350 kHz for <sup>7,11</sup>Be<sup>+</sup> and 1 MHz for <sup>10</sup>Be<sup>+</sup>. In this case equation 6.20 leads to the final error for the isotope shift:

$$\Delta \nu_{\rm IS}^{A,A'} = \sqrt{\left(\Delta \nu_0^{\rm A}\right)^2 + \left(\Delta \nu_0^{\rm A'}\right)^2 + \left(\Delta \nu_{\rm rc}^{\rm A} - \Delta \nu_{\rm rc}^{\rm A'}\right)^2}.$$
 (6.21)

The interval A factors are treated differently, because these are a direct fitting result in collinear and anti-collinear spectra. For all results of one isotope the fitting errors  $\Delta A$ of the A factors are weighted by the corresponding fit uncertainty (reduced  $\chi^2$ ).

$$\Delta A^w = \Delta A \cdot \sqrt{\chi^2_{\rm red}} \tag{6.22}$$

The A factors of all scans of an isotope were plotted with their errors  $\Delta A^w$  and a  $\chi^2$ -fit of a constant  $A^{\text{mean}}$  was performed, as shown in figure 6.13. The resulting fit error and  $\chi^2_{\rm red}$  are used in a next step to calculate the final error of the mean value analog to equation 6.22. An additional error, which arises from the voltage amplification of the DAC value by the Kepco, which would linearly shift the A factor value, must also be included and is quadratically added to the fitting error.



Figure 6.13: Fit of all determined A factors in the 2s state of  ${}^{9}\text{Be}^{+}$  with a constant. Mean value and reduced  $\chi^{2}$  are indicated in the plot.

The Kepco calibration measurements during the off-line beam time could not be used for data analysis, because of problems with the data acquisition system. Instead the Kepco calibration of previous experiments was used. This introduced an additional uncertainty but the contribution to the uncertainty of the determination of the absolute transition frequency is only in the order of some kHz. The problem of the data acquisition system was fixed before the on-line beam time. The Kepco calibration will be introduced and discussed in section 6.7.

#### 6.6.3 Recoil correction

The recoil correction has to be included in the data analysis and the determination of the absolute transition frequency. This effect should not be mixed up with the error arising from recoil shifts and has to be included. For an ion with mass  $m_0$  at rest that absorbs a photon, energy and momentum conservation

$$h \cdot \nu_{\text{photon}} = \frac{1}{2}m_0c^2 + h\nu_0$$
 (6.23)

$$m_0 \cdot v = \frac{h \cdot \nu_{\text{photon}}}{c} \tag{6.24}$$

with the photon energy  $h \cdot \nu_{\text{photon}}$  and the real resonance frequency  $\nu_0$  must be fulfilled. Hence, the atom will have a velocity v after the interaction and the additional kinetic energy must be carried by the incoming photon. Solving the equation for the real transition frequency leads to :

$$\nu_{\text{correction}} = \frac{h\nu_{\text{photon}}^2}{m_0 c^2}.$$
(6.25)

In the case of  ${}^{9}\text{Be}^{+}$  this effect contributes with 226 kHz to the total transition frequency and below 200 kHz for  ${}^{11}\text{Be}^{+}$ .

	Off-line beam time	Literature	Source
	MHz	MHz	
$oldsymbol{ u}_0$	$957199552.3_{-1.0}^{+0.8}$	957199652(120)	[BOL85]
		957199365(240)	[PUC08]
		957199365(150)	[YAN08]
$\mathbf{A}_{2s}$	-624.97(04)	-625.008837048(10)	[WIN83]
$\mathbf{A}_{2p}$	-117.99(04)	-118.6(3.6)	[BOL85]
		-117.932(3)	[PUC09]

Table 6.3: Absolute transition frequency in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition of <sup>9</sup>Be<sup>+</sup> and the hyperfine splitting constants  $A_{2s}$  and  $A_{2p}$ .

## 6.6.4 Transition frequency and hyperfine splitting of <sup>9</sup>Be<sup>+</sup>

Figure 6.14 shows a histogram of analyzed data, which has been recorded at an acceleration voltage of 60 kV. The mean value for the absolute transition frequency  $\nu_0$  of the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition is

$$\nu_0 = 957199552.3^{+0.8}_{-1.0} \text{ MHz} \tag{6.26}$$

where the uncertainty is calculated as depicted in section 6.6.2. The uncertainty for the measured absolute transition frequency is asymmetric, because of the recoil effect that shifts the observed transition frequency only to higher frequencies. The arising error is quadratically added to the statistical uncertainty of 800 kHz and defines the lower border of the total uncertainty. Comparison with literature values as listed in table 6.3 shows good agreement within the uncertainty and an improvement in the accuracy by about 2 orders of magnitude for the absolute transition frequency and the hyperfine splitting constant  $A_{2p}$ . The good agreement indicates that the Kepco factor from the previous beam time must be very close to the value of the off-line beam time since the A factor is directly sensitive to the Kepco calibration. The experimental approach seems to be rather insensitive to experimental conditions within the boundary of careful preparation. Only the alignment



Figure 6.14: Histogram of all transition frequencies of <sup>9</sup>Be measured at 60 kV acceleration voltage in the off-line beam time. Bars on the top with numbers one to four indicate the contributions of different uncertainties to the overall error as discussed in the text.

of the laser beams contributes with approximately 500 kHz to the total uncertainty. The contribution of these measurements cause the increased probability at higher frequencies in the histogram. Note that the lower points in figure 6.11 (b) are completely responsible for the higher frequency contribution on the left side of the histogram in figure 6.14. This figure also illustrates that the error estimation is rather conservative. In the upper part of figure 6.14 all contributions of the discussion of uncertainties in section 6.6.2 are plotted. The numbers correspond to the uncertainty  $\Delta \nu_0^A$  calculated by:

1. 
$$\Delta \nu_0^A = \text{sem}$$

2. 
$$\Delta \nu_0^A = \sqrt{\mathrm{sem}^2 + \Delta \nu_{\mathrm{align}}^2}$$

3. 
$$\Delta \nu_0^A = \sqrt{\mathrm{sem}^2 + \Delta \nu_{\mathrm{align}}^2 + \Delta \nu_{\mathrm{fc}}^2}$$

4. 
$$\Delta \nu_0^A = \sqrt{\operatorname{sem}^2 + \Delta \nu_{\operatorname{align}}^2 + \Delta \nu_{\operatorname{fc}}^2 + \Delta \nu_{\operatorname{rc}}^2}$$

## 6.6.5 Laser spectroscopy for voltage calibration

Another possibility that arises from collinear/ anti-collinear laser spectroscopy and the determination of absolute transition frequencies is the voltage calibration of the ISOLDE high voltage dividers. From the observed transition frequencies one can calculate back to the expected kinetic energy of the ions and compare it to the measured voltages.

Both ISOLDE voltage dividers ASTEC-1 and ASTEC-2 were analyzed and voltage deviations of 39(1) V for ASTEC-2 and -15(1) V for ASTEC-1 were found. This means that the measured voltage at ASTEC-2 is too high, while it is too low for ASTEC-1. Those measurements were confirmed with a high voltage divider, which was developed in the KATRIN collaboration, as described in more detail in [KRI08]. The results of these calibrations have been included in the acceleration voltages used during data analysis. It should be mentioned, that this result partially resolved discrepancies that were observed in previous COLLAPS beam times on Hg and Cu. There, inconsistent isotope shifts were observed in measurements with ASTEC-1 and ASTEC-2 [YOR08].

## 6.7 Results of the on-line beam time

Spectra of the stable and radioactive isotopes were taken during the on-line beam time similarly as for  ${}^{9}\text{Be}^{+}$  in the off-line beam time. Typical spectra as they were obtained for  ${}^{7,9,10,11}\text{Be}^{+}$  are shown in figure 6.15 and are analyzed as described in the previous section. Each spectrum for collinear and anti-collinear direction is the sum of at least 50 and for  ${}^{11}\text{Be}$  at least 400 scans. The number of steps per scan was chosen between 400 or 800 channels, which results in a resolution between 80 kHz and 160 kHz. The signal accumulation time of the photomultiplier had to be changed in some cases from 10 to 20 ms, when a 50 Hz ripple appeared on the photomultiplier signal as well as on wire scanner signals in front of the COLLAPS beam line, which can probably be attributed to the ion source potential. This ripple might be the reason of the "bunching" effect that is



Figure 6.15: Spectra of  $^{7,9,10,11}$ Be<sup>+</sup> obtained during the on-line beam time. Experimental data (blue) and fitted lineshapes (red).



Figure 6.16: Kepco calibrations of the on-line beam time. • is the Fluke offset voltage during the calibration measurement while the black • show the Kepco amplification factor of the corresponding calibration measurement.

observed for the data points along the lineshape of  ${}^{10}\text{Be}^+$ . It exhibits some peculiarities which will be discussed in the next section.

## 6.7.1 Calibration of the Kepco voltage amplifier

A crucial point for data analysis is the knowledge of the applied voltages. This is less severe for the approach described in this thesis, but particularly the extraction of the A factors is dependent of the applied voltages. For this reason the amplification factor of the Kepco amplifier has to be measured several times during a beam time. In those calibration measurements the DAC voltage is slowly scanned from -10 to 10 Volt and the voltage at the optical detection region is measured with the Prema digital voltmeter. Approximately 48 data points were recorded with an integration time of 4 s. The insert in figure 6.16 shows the result of such a Kepco calibration and the fitted linear function. The slope of such a linear regression line is plotted in the figure for 6 calibrations that were performed during the beam time. The points scatter slightly around a mean value of

$$K_{\text{Kepco}} = 50.4124(32). \tag{6.27}$$



Figure 6.17: Laser power dependence of the observed transition frequency of <sup>10</sup>Be<sup>+</sup>.

The scattering of  $K_{\text{Kepco}}$  seems to be correlated with the applied Fluke voltage. A large Fluke voltage coincides with smaller, a small Fluke voltage with a larger Kepco factor. However the correlation is weak and due to the long time between the measurements, there are many other possibilities. Even though the amplitude of the variation is so small that it does not matter for the isotope shift measurements it should be tested by time.

## 6.7.2 Systematic effects for <sup>10</sup>Be

The line shape of <sup>10</sup>Be as illustrated in figure 6.15 reveals a non trivial line shape that exhibits a slight asymmetry close to the resonance. The reason for this distorted line profile is not completely understood, yet. The peak has usually comparable asymmetries in collinear and anti-collinear spectra, which shifts the peak to higher voltages in both cases. But since this means in the frequency domain a shift in opposite directions, the geometric average of the two shifted frequencies should be rather insensitive to this effect. It is estimated that the remaining frequency shift is the order of some 10 kHz which is much smaller than other systematic effects.

<sup>10</sup>Be is the only even isotope that was investigated during the on-line beam time. It has no nuclear spin and therewith no hyperfine splitting. As a result uncertainties which are introduced by recoil effects and the higher scattering rate have to be discussed in more detail. Measurements of <sup>10</sup>Be were performed at different collinear and anti-collinear laser powers. The obtained absolute transition frequencies are plotted in figure 6.17 against the power of the two laser beams. All data points are fitted with a plane, which crosses the frequency axis for vanishing laser powers at a frequency of:

$$\nu_0^{\text{fit}} = 957216875.1 \text{ MHz},\tag{6.28}$$
which is 1.7 MHz below the averaged value  $\overline{\nu_0}$  of all measurements as listed in table 6.4. If the interaction with the photons shifts the resonance line, one would expect a similar effect in both directions. But contrary to the asymmetry of the lineshape, this effect would shift the line in both cases in the same direction in the frequency domain: Collinearly the ions are accelerated by photon absorption, hence a lower acceleration voltage is required to come into resonance, whereas anti-collinearly the ions are decelerated and higher acceleration voltages have to be used. In the frequency domain this shifts the resonance in both cases to higher frequencies. However, according to figure 6.17 an effect on the absolute transition frequency as a function of the laser power is only observed in collinear, but not in anti-collinear direction. Hence, it is not absolutely clear whether the observed behavior is indeed due to the photon recoil. Nevertheless, it cannot be totally excluded and a possible systematic shift must be added to the total uncertainty. Typical laser powers of 3 to 4 mW have been used in the measurements which leads to a contribution of 1.2 MHz. The uncertainty at this powers is calculated by using the bigger slope of the plane, which is 300 kHz/mW and agrees well with the assumptions of section 6.6.2. Even if this uncertainty contributes with only 600 kHz to the isotope shift measurement an error of 1 MHz is estimated, which is quite conservative, as the power variation of the anti-collinear laser does not seem to have any influence.

### 6.8 Results and discussion

#### 6.8.1 Absolute transition frequencies

The results for all measurements, which have been performed in the off- and on-line beam time are summarized in table 6.4. The total error  $\Delta\nu_0$  is separated into a symmetrical part  $\Delta\nu_0^{\text{sym}}$  and the recoil part  $\Delta\nu_0^{\text{recoil}}$ . The first part  $\Delta\nu_0^{\text{sym}}$  includes the statistical uncertainty (sem), the alignment uncertainty ( $\Delta\nu_{\text{align}}$ ) and the contribution of the frequency comb ( $\Delta\nu_{\text{fc}}$ ) which were added quadratically. The second part  $\Delta\nu_0^{\text{recoil}}$  is the estimated shift of the transition frequency due to multiple photon absorption and is quadratically added to  $\Delta\nu_0^{\text{sym}}$ . It shifts the frequency systematically to higher frequencies and therewith the arising uncertainty contributes only in negative direction.

The absolute transition frequencies were determined for the first time with the denoted accuracy of approximately 1 MHz. Previous measurements for  ${}^{9}\text{Be}^{+}$  [BOL85] had an uncertainty of 120 MHz, while the transition frequencies for other isotopes were only derived by mass shift calculations. The experimental values of the presented experiment agree within the errors with these results, while the accuracy is increased by two orders of magnitude. Ongoing improvements in theory will allow the calculation of the nuclear charge radii directly from absolute transition frequencies in far future. As both parameters, the absolute transition frequency as well as the nuclear charge radius, were measured in this experiment with the needed accuracy they will provide theoretical physicists with essential test parameters.

Table 6.4: Absolute transition frequencies for <sup>7,9,10,11</sup>Be<sup>+</sup> and their contributing uncertainties. The symmetrical part  $\Delta \nu_0^{\text{sym}}$  is listed separately from the uncertainty that includes the recoil effect which always shifts the observed transition frequencies to higher frequencies. N is the number of analyzed spectra. The description of the listed uncertainties can be found in the text.

Α	Run	Ν	$\overline{oldsymbol{ u}_0}$	sem	$\Delta \nu_{\rm align}$	$\Delta \nu_{\rm fc}$	$\Delta \nu_{\rm rc}$	$\Delta \nu_0^{ m sym}$	$\Delta \nu_0^{ m recoil}$
			MHz	MHz	MHz	MHz	MHz	MHz	MHz
9	off-line	88	957 199 552.3	0.01	0.5	0.58	0.7	$\pm 0.8$	-1.04
7	on-line	21	957 150 316.1	0.15	0.5	0.58	0.9	$\pm 0.8$	-1.19
9	on-line	21	957 199 553.0	0.35	0.5	0.58	0.7	$\pm 0.8$	-1.04
10	on-line	48	957 216 876.9	0.21	0.5	0.58	1.26	$\pm 0.8$	-1.49
11	on-line	11	957 231 118.1	0.34	0.5	0.58	0.57	$\pm 0.8$	-1.01

#### 6.8.2 A factors and magnetic moments

All obtained A factors for the ground and excited state in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition were analyzed as described in section 6.6.2 and are listed in table 6.5. The observed A factors show excellent agreement with previous measurements, while their accuracy could be increased in almost all cases. It should be noted that the literature values for the A factors of <sup>7,11</sup>Be as listed in table 6.5 are calculated from measurements of their magnetic moments by  $\beta$ -NMR experiments and optical hyperfine measurements. Also theoretical predictions of the  $A_{2p}$  value by [PUC09] agree well with the presented data. The A factor determination can be used to extract the magnetic moments for odd isotopes with reference to <sup>9</sup>Be. High precession measurements of the A factor in the ground state  $A_{2s} = -625.008837048$  MHz [WIN83] and the corresponding nuclear magnetic moments  $\mu = -1.177432(3) \mu_N$  [ITA83] allow the determination of the nuclear magnetic moments for <sup>7,11</sup>Be as listed in table 6.5 by

$$\mu_I \left( {}^{A} \mathrm{Be} \right) = (1 + \Delta_{A,9}) \frac{A \left( {}^{A} \mathrm{Be} \right)}{A \left( {}^{9} \mathrm{Be} \right)} \cdot \frac{I \left( {}^{A} \mathrm{Be} \right)}{I \left( {}^{9} \mathrm{Be} \right)} \cdot \mu_I ({}^{9} \mathrm{Be})$$
(6.29)

The contribution of the hyperfine anomaly  $\Delta_{A,9}$  is in the order of  $10^{-5}$  [OKA08] and is neglected. The magnetic moment of <sup>7</sup>Be was measured previously at COLLAPS using fluorescence detection [KAP98]. The new value agrees well with the old result, but the accuracy has been improved by more than one order of magnitude. Recently the group at RIKEN reported a new precise measurement of the <sup>7</sup>Be hyperfine splitting which is about one order of magnitude more precise than the measurement presented here since it has been measured by optical-microwave double-resonance technique. But it is still in excellent agreement with the presented values. The magnetic moment for <sup>11</sup>Be, obtained in  $\beta - NMR$  experiments could be confirmed.

Α		$A_{2s}$	$A_{2p}$	$\mu$	Source	
		MHz	MHz	$\mu_{ m N}$		
9	off-line	-624.971(045)	-117.999(041)		this work	
	on-line	-625.001(149)	-118.028(082)		this work	
		-625.008837048(10)	-118.6(3.6)	-1.177432(3)	[WIN83, BOL85]	
			-117.932(3)		[PUC09]	
7	on-line	-742.984(246)	-140.188(178)	-1.3995(5)	this work	
		-742.772	-140.8	-1.398(15)	[KAP98]	
			-140.157(3)		[PUC09]	
		-742.77228(43)		-1.39928(2)	[OKA08]	
11	on-line	$-2\ 677.665(774)$	-505.470(499)	-1.6813(5)	this work	
		-2 677.306	-508.1	-1.6816(8)	[GEI99]	
			-505.245(16)		[PUC09]	

Table 6.5: Obtained  $A_{2s}$  and  $A_{2p}$  values for <sup>7,9,10,11</sup>Be<sup>+</sup> during off- and on-line beamtimes as well as derived magnetic moments and comparison with previous measurements and theoretical results.

#### 6.8.3 Isotope shifts and nuclear charge radii

The results for the isotope shifts between <sup>7,10,11</sup>Be<sup>+</sup> relative to <sup>9</sup>Be<sup>+</sup> and the resulting nuclear charge radii are summarized in table 6.6. The absolute transition frequency for <sup>9</sup>Be<sup>+</sup> from the off-line beam time is not included. Within its error it agrees with the results of the on-line beam time and does not need to be treated separately. Including the data of the mass shift (tab. 3.1) from [PUC08], the field shift  $\nu_{FS}$  can be directly extracted from the derived isotope shifts. Taking the charge radius of <sup>9</sup>Be<sup>+</sup> [JAN72] as reference leads finally to the root mean square nuclear charge radius of all investigated isotopes.

The determined nuclear charge radii as listed in table 6.6 are plotted in figure 6.18 and compared to theoretical predictions. Isotope shifts in the D2 line were also measured and the extracted charge radii, which are less accurate due to the unresolved hyperfine structure, agree with the presented data [ZAK09]. The nuclear charge radii decrease from <sup>7</sup>Be to <sup>10</sup>Be and increase again for <sup>11</sup>Be. This behavior can probably be explained by clustering effects as illustrated in figure 6.19. <sup>7</sup>Be is supposed to be formed by an  $\alpha$  and a helium-3 cluster, while the next heavier isotope <sup>8</sup>Be would consist of two  $\alpha$  particles but is not bound. <sup>9,10,11</sup>Be are two  $\alpha$  particles which are bound with one, two or three neutrons respectively. Theoretical predictions for the charge radii of these isotopes show generally good agreement with the experimental results. This is in contrast to the case of Li isotopes [SAN06a].

The increase of the nuclear charge radius of <sup>11</sup>Be can be attributed either to the motion



Figure 6.18: Experimental charge radii of beryllium isotopes from isotope-shift measurements (•) compared with values from interaction cross-section measurements (•) and theoretical predictions: Greens-Function Monte-Carlo calculations (+) [PIE01, PIE02], Fermionic Molecular Dynamics ( $\Delta$ ) [TOR08], ab-initio No-Core Shell Model ( $\Box$ ) [FOR05, NAV06, NAV08].



Figure 6.19: This plot illustrates the supposed clustering of <sup>7</sup>Be (upper left) to <sup>11</sup>Be (lower right). The red square indicates that <sup>8</sup>Be is an unbound nucleus. (<sup>7</sup>Be-<sup>10</sup>Be: courtesy of P. Müller)

Table 6.6: Absolute transition frequency  $\nu_0$ , isotope shift  $\nu_{IS}$ , field shift  $\nu_{FS}$  and change in the mean square nuclear charge radius  $\langle \delta r^2 \rangle$  as well as the nuclear charge radius  $r_c$  for all investigated isotopes. The relative uncertainty  $\Delta r/r$  of the obtained nuclear charge radii is below 1 percent.

Α	$\nu_0$	$ u_{IS} $	$ u_{FS} $	$\langle \delta r \rangle^2$	r	$\Delta r/r$
	MHz	MHz	MHz	$\mathrm{fm}^2$	${ m fm}$	
7	$957 \ 150 \ 316.09(52)$	$49\ 236.9(9)$	11.05(9)	0.66(5)	2.647(17)	$6.10^{-3}$
9	957 199 55 $3.03(61)$	0	0	0	2.519(12)	
10	$957\ 216\ 876.86(54)$	-17 323.8(13)	-13.3(13)	-0.79(08)	2.357(18)	$9.10^{-3}$
11	957 231 118.07(60)	-31 565.0(9)	-4.9(9)	-0.29(05)	2.463(16)	$7.10^{-3}$

of <sup>10</sup>Be relative to the center of mass which changes due to the additional neutron, or to an increasing distance between the two  $\alpha$  clusters. In the first case the change of the center of mass between <sup>10</sup>Be and <sup>11</sup>Be can be directly assigned to the nuclear charge radii according to

$$r_{\rm c}^2(^{11}{\rm Be}) = R_{\rm cm}^2 + r_{\rm c}^2(^{10}{\rm Be}).$$
 (6.30)

The center of mass in <sup>11</sup>Be is shifted by approximately 0.7 fm, which leads to a distance of 7 fm between the neutron and the  ${}^{10}$ Be core and is illustrated in figure 6.20 (A). This is in good agreement with calculations of [JON87]. They calculated the distance between halo neutron and the<sup>10</sup>Be core based on measurements of the one neutron separation energy of 0.5 MeV in 1987. The used model was simple: A single neutron is assumed to be bound in an s-orbit in a Woods-Saxon potential and the potential depth is adjusted such that the neutron separation energy is reproduced. Then the wavefunction of the neutron exhibits an exponential decay length of  $\sim 7$  fm. If the change in the charge radius is solely attributed to a change in the charge radius of the <sup>10</sup>Be core, the change in the distance between the two  $\alpha$  clusters can be extracted. This is illustrated in figure 6.20 (B), where results of Fermionic Molecular Dynamics calculations for <sup>10</sup>Be and <sup>11</sup>Be are shown. For <sup>10</sup>Be the  $\alpha$  particles are separated by about 1.65 fm. An increase of about 10 % to 1.8 fm is required to explain the  $\langle \delta r \rangle^2$  between <sup>10</sup>Be and <sup>11</sup>Be. This model is certainly simplified too much since some part of the center of mass motion must be present in a one-neutron halo and the truth lies most probably between these two extremes. Isao Tanihata proposed to combine the information of the nuclear charge radius, the nuclear matter radius and the B(E1) measurement of <sup>11</sup>Be to determine the neutron-core distance more accurate.

All theoretical predictions, which will be roughly explained in the following paragraph [SAN06b], show in principle the same trend. At first the decrease of the nuclear charge radius from <sup>7</sup>Be to <sup>10</sup>Be and then the increase for <sup>11</sup>Be. For <sup>9</sup>Be and <sup>10</sup>Be the agreement with Greens-Function Monte Carlo calculations is striking. These calculations are currently the most fundamental tool for the ab-initio calculation of nuclear structure. This model is based on individual nucleons that interact via two-nucleon and three-nucleon forces. Two nucleon potentials are obtained from nucleon-nucleon scattering, whereas three-nucleon potentials are adapted by global adjustment of potential parameters by



Figure 6.20: (A) Center of mass movement in <sup>11</sup>Be under the assumption of an inert <sup>10</sup>Be core. In this case, the center of mass in <sup>11</sup>Be is located approximately 0.7 fm off the center of gravity of the <sup>10</sup>Be core. This causes the core to orbit around the center of mass and due to the mass ratio the halo neutron must be at a distance of 7 fm. (B) The proton and the neutron distributions  $\rho_{N,P}$  in one intrinsic state of the FMD calculations are plotted for <sup>10,11</sup>Be [NEF09]. It illustrates the clustering of these nuclei and the appearance of the neutron halo in <sup>11</sup>Be. A change in the cluster distances R1 and R2 respectively would also contribute to  $\delta \langle r^2 \rangle^{9,10}$ .

comparing the binding energy of ground and excited states for a large number of light nuclei. Calculations become exponentially more complex with an increasing number of nucleons and <sup>11</sup>Be has not been modelled so far.

Ab-initio Large-Basis No-Core Shell Model (LBNCSM) calculations, treat a nucleus as a system of A nucleons, which are interacting by realistic internucleon forces. For the calculation a large but finite number of harmonic-oscillator basis functions is used. The constricted number of basis makes it necessary to use an effective interaction, which is calculated based on the internucleon interaction that is appropriate for the employed basis. Recent results of this method are included in the graph and reproduce the trend along the isotopic chain fairly well. The change between <sup>7</sup>Be to <sup>9</sup>Be shows even the best agreement of all theoretical models.

In the Fermionic molecular dynamics (FMD) model [NEF05], the nucleon-nucleon interaction is derived from the Argonne V18 interaction. The change in the charge radius from <sup>9</sup>Be to <sup>11</sup>Be is extremely well reproduced, whereas <sup>7</sup>Be appears a little bit to compact.

It should be mentioned that most nuclear theories treat the protons and neutrons as point-like particles and extracted nuclear charge radii are therefore reported as pointproton  $\langle r_P^2 \rangle$  and point-neutron  $\langle r_N^2 \rangle$  radii. For comparison with measured charge radii the root-mean-square charge radii of proton and neutron have to be included. The mean square charge radius of the proton  $\langle R_P^2 \rangle = 0.895(18) \text{ fm}^2$  [SIC03] and of the neutron  $\langle R_N^2 \rangle = -0.113(7) \text{fm}^2$  [KOP95] are folded into these results according to:

$$r_C^2 = \left\langle r_P^2 \right\rangle + \left\langle R_P^2 \right\rangle + \frac{N}{Z} \left\langle R_P^2 \right\rangle + \frac{3\hbar^2}{4M_p^2 c^2} \tag{6.31}$$

with the Darwin-Foldy correction  $\frac{3\hbar^2}{4M_p^2c^2} \sim 0.033 \text{ fm}^2$ .

# Chapter 7

# Towards laser spectroscopy at ISOLTRAP

The measurements presented in the previous chapter yielded nuclear charge radii that have an accuracy of approximately 1%. For nuclear structure theory that is currently more than sufficient because theory will not offer higher accuracy in the next time. Isotope shift measurements in an rf-trap laser-cooled ions can provide results with an accuracy that is almost one order of magnitude larger. Those measurements can be used for validating the presented data and give access to further interesting properties:

- Determination of the magnetic moments by rf-optical double resonance spectroscopy. This will allow a precision determination of the hyperfine anomaly and therewith further information about the nuclear structure.
- Comparison of the isotope shift in the D1 and D2 line. This allows the determination of the so-called splitting isotope shift. It can be very well predicted by atomic theory since the mass independent term as well as the finite size terms do cancel. However, so far this splitting isotope shift could not be determined with high accuracy in lithium like systems. In the case of Li this effect is very small and inconsistent and contradicting measurements exist. In Be<sup>+</sup> the effect is much larger and can be determined by collinear laser spectroscopy but with limited accuracy [ZAK09].

Hence it is still an interest in the high resolution measurements of trapped and laser cooled Be<sup>+</sup> ions as they were proposed and are also planned and performed at RIKEN [OKA08]. In this chapter the developments and preparations towards trapping of radioactive Be<sup>+</sup> isotopes at ISOLDE by using the existing ISOLTRAP facility that will lead in the future to such high accuracy measurements, are described.

## 7.1 Planed experimental setup at ISOLDE

Figure 7.1 shows a rough outline of the ISOLTRAP setup, which is used for high precision mass measurements in Penning traps. The ion beam from the ISOLDE front-end as it was described in the previous chapter is guided into a radio frequency cooler and buncher



Figure 7.1: Be ions from the ISOLDE front-end are transported to the ISOLTRAP facility. Here, they can be accumulated and cooled in the RFQ structure and extracted to ground potential. A pulsed drift tube is used for reduction of the kinetic energy to approximately 2500 eV, which is suitable for ion confinement in Penning traps. For Be spectroscopy the ions will pass the 90° bender and enter a second pulsed drift tube, where their kinetic energy can be reduced to a few eV.

(RFQ) and extracted with a kinetic energy of approximately 2.5 keV. An electro static 90° bender is used to transport this extracted ion bunch into a first Penning trap for beam purification and finally into the Penning trap for high precision mass measurements.

Compared to the ISCOOL RFQ, which has recently be installed at the ISOLDE general purpose mass separator (GPS), the ISOLTRAP facility provides the advantage that Be ions can not only be cooled and bunched in the RFQ but also their kinetic energy can be decreased to a defined level in a pulsed drift tube (PDT). The operation of this device will be discussed in section 7.1.2. Hence, it was concluded that the best place at ISOLDE to install this experiment is behind the ISOLTRAP RFQ. Instead of guiding the ions to a vertical tower of Penning traps, the 90° bender will be removed or replaced with a four way quadrupole deflector and the ions will be transported straight through this section as indicated in figure 7.1. An additional PDT is used for further beam energy reduction as well as ion optics to guide the ion beam into the linear Paul trap. Because laser access along the optical axis of the Paul trap is needed from both sides for beam alignment and spectroscopy also an electrostatic 90° bender has to be inserted.

#### 7.1.1 The Isoltrap Cooler and buncher

The RFQ at ISOLTRAP was developed between 1998 and 2001 [HER01a] and was installed in the existing ISOLTRAP setup to increase the overall efficiency and to allow mass measurements of very short-lived, rare isotopes. With this additional device it was possible to measure for example  ${}^{33}\text{Ar}^+$  with a half life of only 174 ms in a Penning trap for the first time. The principle of operation is as follows: ions are confined in a linear Paul



Figure 7.2: Left: Photo of the cooler and buncher at ISOLTRAP. Right: A schematic of the cooler and buncher with the applied DC potential for longitudinal ion confinement [HER01b]. The buffer gas cooled ions accumulate in the potential minimum.

trap, cooled by buffer gas collisions, thermalized ions are accumulated in the potential minimum of the trap and finally extracted as a short ion bunch.

The RFQ is described in detail in [HER01a, HER01b] and only a brief overview will be given here. Figure 7.2 shows a photo and a schematic of the cooler and buncher. It has a length of 0.81 m and a field free radius of 6 mm. The quadrupole rods are segmented into 26 parts, to which different DC potentials for longitudinal ion confinement can be applied. The Kinetic energy of the ion beam that enters the RFQ is approximately 30 to 60 keV. This energy is electrostatically reduced to some eV by putting the complete RFQ on a high voltage platform, which allows ion confinement in the quadrupole and electrostatic potential of the RFQ. Trapped ions dissipate their radial and longitudinal kinetic energy in the buffer gas due to collisions as illustrated in figure 7.3. This process takes a few milliseconds depending on the cooling gas pressure (typically  $10^{-2}$  mbar of He), the ion's kinetic energy and the



Figure 7.3: Simulation for the energy dissipation of  ${}^{39}\text{K}^+$  in longitudinal (z) and radial (r) direction in presence of  $10^{-2}$ mbar He and an incident energy of 10 eV.

coolant mass [HER01b]. Best cooling results can be achieved, when the ion's mass is much higher than that of the coolant gas, because the ion experiences in this case a viscose damping force. This is the reason, why cooling of Be<sup>+</sup> in He-buffer gas is not suitable. For a more efficient cooling process the coolant has to be  $H_2$ , which is a non trivial change in the experimental setup due to safety restrictions and is still under preparation. Once



Figure 7.4: Ions that are leaving the RFQ are first accelerated into the pulsed drift-tube. Their kinetic energy is then determined by the potential difference between RFQ and the tube. If the potential of the pulsed drift-tube is switched to ground, while the ions are inside, the ions keep their initial energy once they left the tube.

the ions are cooled down to thermal equilibrium with the buffer gas, they can be extracted to ground potential by switching the DC potentials for longitudinal ion confinement to appropriate voltages as indicated in figure 7.2. The emittance and time profile of the released ion bunch does strongly depend on the buffer gas temperature as well as on the trapping potentials. It has a typically puls width of only a few  $\mu s$  and very low emittance.

Cooling and bunching of ion beams in those RFQ devices is nowadays widely used in radioactive beam facilities for improvements in beam emittance and sensitivity [WIE06, CAM02, FRA08].

#### 7.1.2 Pulsed drift tube

The extraction of the ion bunch to ground potential is not suitable for a transfer into a Penning trap. At ISOLTRAP an elegant way to modify the ion's kinetic energy from, e.g. 60 keV to 2.5 keV after extraction is used. The ions are released from the buncher and enter a PDT through beam shaping optics. This is a 38 cm long metal tube, which is connected to a fast high voltage switch. The potential between the RFQ and the PDT defines the ion's final kinetic energy. When the ion bunch passes the middle of the drift tube (field free region) the potential is set to ground potential and the ions enter the last part of the beam line at ground potential with the initial kinetic energy (see figure 7.4).

With this simple operational procedure it is not possible to achieve a high transmittance for a final kinetic energy of less than 2.5 keV, because the pulsed drift tube also acts as a strong ion optical element, which becomes dominant for low kinetic energies. The transmittance is also limited, because the ions have to pass through the magnetic fringe fields of the Penning traps of ISOLTRAP and are disturbed due to the Lorentz force.



Figure 7.5: Simulation of Be<sup>+</sup> ions with an initial kinetic energy of 2.5 keV which are decelerated to 10 eV in a PDT. On the left, the simulation array is shown. The ion parameters are recorded in plane 1 and 2. On the right, the result is shown. The x axis is the initial high-voltage x of the PDT, which is switched to x - 2490 V to obtain a final kinetic energy of 10 eV for the ions.

Hence, it was chosen to extract the ions from the PDT with about 2.5 to 3 kV and to reduce the ion's kinetic energy further in a second PDT to some eV. To retard the ions from an initial energy of e.g. 2.5 keV to an energy of 10 eV the potential of the PDT can be switched from 2.49 keV to ground potential or from ground potential to -2.49 keV. In both cases the ions will have a final energy of 10 eV but very low transmission, because the ion beam is strongly defocused either at the exit or at the entrance of the PDT. Simulations show that the optimal transmission through a PDT strongly depends on the initial and final potential of the PDT, as shown in figure 7.5. A transmittance of almost 100 percent can be achieved by switching the potential from +1500 to -990 V in the case discussed here. Based on these considerations, the second PDT has been designed as a 40 cm long CF-40 tube in a PEEK housing, which is isolated with 10 cm long ceramic tubes against the rest of the vacuum chamber and the beam line. The high voltage switch must be capable to switch between those two voltages much faster than the transit time of <sup>7</sup>Be ions through the PDT. Such a fast voltage switch was produced and successfully tested as shown in figure 7.6. Depending on the experimental requirements one can choose between two options which can be easily adjusted inside the electronic box. Either one can perform a fast switch from low to high, or from high to low potential. The required circuit board layouts for both cases are illustrated in figure 7.6 (A) and (B). Figure 7.7 shows a photo of the PDT in the PEEK housing and the switching time between two high-voltages that was measured with a high voltage probe. The observed switching time was less than 200 ns and is approximately 10 times faster than the transit time of 1.5  $\mu$ s for  ${}^{7}\text{Be}^{+}$  with a kinetic energy of 2.5 keV. More precise measurements of the rise time could not be performed, because the used high-voltage probes had a time constant of about 200 ns, which limits the time resolution. The attainable ion puls duration for Be<sup>+</sup> in the RFQ is not known yet, but typical puls widths of a few  $\mu$ s will limit the overall efficiency, because it is larger than the transit time. But because of limited space for the



Figure 7.6: Photo and circuit diagrams of the high-voltage switch. (A) is the circuit diagram for fast switching from low to high potential, while (B) is used for fast switching from high to low potential.

experimental setup at ISOLTRAP a longer PDT could not be used and the production rates are sufficiently large for all isotopes so that losses of a factor of 2 do not matter since - in the end - single ion spectroscopy will be performed.

### 7.1.3 Ion optics

The ion beam has to be efficiently guided from the PDT into the ion trap. For this purpose intensive simulations with SimIon 8.0 were performed, to find the best suitable geometry for the ion optical elements and appropriate voltages for these elements. The simulations have been performed for a  ${}^{9}\text{Be}^{+}$  beam with a kinetic energy of 3 keV and an approximated beam emittance with a beam diameter of 6.4 mm and an opening angle of 0.6 deg, which corresponds approximately to the measured beam emittance of ISOLTRAP's RFQ at a



Figure 7.7: Photo of the second PDT, which is used to reduce the ion's kinetic energy to some eV. On the right hand side the reachable switching times are shown. Slow and fast slope can be changed.

position 50 cm behind the 90 degree bender [HER01b].

This ion beam is decelerated in the second PDT to 10 eV by switching it from 1770 V to -1220 V. Figure 7.8 shows an illustration of the experimental setup, which was built based on the simulation results. The ion optical setup consists of a skimmer to reduce the contamination of the vacuum chamber with radioactive isotopes, two einzellenses for beam focusing and a 90° bender with three additional correction electrodes. Also a dummy trap, the final trap was not developed at this time of work, at ground potential was inserted to include fringe fields and their influence on the ion trajectories. Figure 7.8 (A) and (B) show the beam profile as well as the beam emittance in the plane, which defines the entrance to the PDT (plane A) and the ion trap (plane B). The transport efficiency from plane A to B is 56 %.

A problem that could arise during the experiment is the electrostatic 90° bender, which acts as an energy filter. Due to this property voltage fluctuations at ISOLTRAP could directly influence the capture efficiency of the trap, because different kinetic energies lead to a different steering in x direction. However, in the COLLAPS measurement it was observed that the ISOLDE voltage is quite stable and should not cause too many problems.

#### 7.1.3.1 Efficiency

With the described setup only some 10 ions/ minute have to be captured in the Paul trap for spectroscopy. ISOLDE delivers a yield of approximately  $10^8$  ions of <sup>11</sup>Be per minute. Therewith, the needed overall efficiency for bunching of the ion beam, decelerating, transfer and trapping is in the order of  $10^{-6}$  or  $10^{-7}$ .



Figure 7.8: Experimental layout constructed based on a series of simulations. The beam profile used for the simulations is shown in (A) and was recorded in the corresponding plane. The ion beam emittance and profile after all ion optical elements is also shown in (B) and was recorded in plane B just in front of the dummy trap.

# Chapter 8

# Tests of the Paul trap

### 8.1 Paul trap

During this work two types of linear Paul traps were developed which are shown in figure 8.1. Type 1 is a *classical* design as described in chapter 4. Because of the limited time which was available for experiments, only this type was tested and will be described in more detail in this section. The design of type 2 is based on experiments, which are performed at the university of Ulm/ Germany, in the department of "quantum information processing" (QIV)". This Paul trap consists of copper plated circuit boards instead of cylindrical rods. The advantage of this design is a high flexibility in the configuration of axial trapping potentials, low manufacturing costs and a very good optical access, while the main disadvantage of this design is a low capture efficiency. A technical drawing of this trap type can be found in appendix A.

Both types are modularly designed, because at a later point rare isotopes as <sup>11</sup>Be<sup>+</sup> should be accumulated in a first big trapping region, where they would be precooled by laser cooling and transported sequentially to a second smaller trapping area, where spectroscopy on single ions



Figure 8.1: The upper photo shows trap type 1, the lower photo trap type 2.

could be performed. The first step to this complex experimental scheme is testing of the ion trap, laser cooling and of course the optical detection with a CCD camera or



Figure 8.2: Illustration of the electrode configuration as used in first test experiments. Further information in the text. All values are given in mm.



Figure 8.3: Pseudo potential for the described trap geometry for  ${}^{9}\text{Be}^{+}$  with an rf amplitude of 200 V<sub>pp</sub> at 13.56 MHz. Right: Complete stability diagram.

a photomultiplier. During this work the ion traps were constructed and trapping was demonstrated by destructive ion detection.

#### 8.1.1 Construction

The trap is made of stainless steel electrodes which are gold plated to reduce stray light reflections from the cooling laser on the electrodes and to prevent possible oxidation. The electrodes have a diameter of 6 mm, which leads in regard to equation 4.4 to a field free radius  $r_0$  of 2.6 mm and lengths of optionally 4.5, 9.5, 14.5, 19.5 or 29.5 mm. They can be assembled in any desirable configuration on a thin metal rod which leads to a total length of 80 mm. All electrodes are separated by small *Macor*<sup>1</sup> pieces that have a length of 0.5 mm and a diameter of 4 mm. The setup as used for experiments concerning laser cooling and optical detection is illustrated in figure 8.2.

The trap is operated with an rf-driving frequency of 13.56 MHz, which can be set

<sup>&</sup>lt;sup>1</sup>Glas ceramic (www.pgo-online.com)

to any amplitude between 0 and 450 V<sub>pp</sub> with a typical operation voltage of 200 V<sub>pp</sub> (q $\approx$  0.1). With these settings a potential depth of approximately 2.2 eV for Be ions can be calculated using equations 4.18 and 4.25. The resulting pseudo-potential that <sup>9</sup>Be<sup>+</sup> ions experience in this trap is plotted in figure 8.3 (left) as well as the stability diagram for the given trap geometry (right). The radial oscillation frequencies are  $\omega_{x,y} = 2.7$  MHz, while the axial oscillation frequency for a trapping potential of 10 V is  $\omega_z = 1$  MHz with a resulting potential depth of approximately 0.25 eV in axial direction.

#### 8.1.2 RF oscillator

The required radio frequency for radial confinement of the ions in the trap is produced in a high voltage tube amplifier. Circuit diagrams of the amplifier and the necessary high voltage power supply can be found in appendix B with a short summary of [LEN07]. Compared to other drivers like simple resonance circuits this approach is more complex, but it has been chosen to provide larger flexibility. Both trap types and different wirings of trap type 1 (compare with figure 4.3) can be tested and, hence, this flexibility of the rf amplifier was desirable, because of different capacitive loads of the devices.

The driving frequency of the amplifier is 13.56 MHz. Depending on the capacitive load the resonance frequency shifts and has to be readjusted with an adjustable capacitor. The input signal is generated in a standard frequency generator and has to be fed into the rf amplifier after pre-amplification to approximately 20 V<sub>pp</sub>, which is required for maximal output performance. The amplifier has two phase-locked outputs which are phase shifted by  $\rho = \pi$ . Both outputs are connected to adjustable capacitors to change the load and therewith the amplitude of each output, while a DC voltage can be applied on each output by a bias-tee. A load of 50 pF on each output allows an rf amplitude of ~500 V<sub>pp</sub> each.

## 8.2 First trapping of Li<sup>+</sup> - destructive

A first test of the ion trap was performed in 2007. At this time, the optical detection system was still not available, hence the ion trap had to be tested with another ion detection method. Ions were created inside the trapping volume by non-resonant ionization of residual gas and Li-atoms with a pulsed Nd:Yag laser. After an adjustable trapping time, ions were released from the trap and were detected with a channeltron<sup>2</sup> - type detector. These experiments, in which the influence of the rf potential, the time of ion creation and the trapping were investigated, demonstrated that ions were trapped but did not support any qualitative information.

#### 8.2.1 Ion source

At this experimental stage Li was used instead of Be to avoid an unnecessary contamination of trap and vacuum chamber with the extremely toxic metal Be. Li atoms were produced in a passively heated stainless steel tube with a diameter of 5 mm

 $<sup>^{2}</sup>$ Electron multiplier device



Figure 8.4: Setup of the trap and the data acquisition electronics, as it was used in first tests. Atoms inside the trapping volume are non-resonantly ionized with a 7 mJ, frequency-trippled Nd:Yag laser puls. After a defined time the trapping electrodes near the channeltron are switched from 90 to 50 V, which releases the ion bunch. Details of the measuring scheme can be found in the text.

and a wall thickness of approximately 0.2 mm as illustrated in figure 8.5. This tube was plugged on a stainless steel rod, which was connected to another rod with a thin

tantalum foil. If a voltage is applied, the tantalum foil gets hot because of the high contact resistance and small cross-section and heats the stainless steel tube. Temperatures of approximately 700°C can be reached with operating currents of 30 to 40 A. The tube has a length of 2 cm and is filled with Li shot, which has a melting point of 180°C. It is mounted 6 cm beside the ion trap and allows the atoms to freely propagate into the trapping volume. Li atoms, which entered the trapping volume were non resonantly ionized with a pulsed, frequency-trippled Nd:Yag laser (355 nm), which was focused with a f=500 mm lens into the ion trap in axial



Figure 8.5: Illustration of the oven used in first trapping experiments to produce Li<sup>+</sup> ions. Applied currents heat the tantalum foil and therewith the Li sample .

direction. A puls duration of <4 ns, a repetition rate of 20 Hz and a puls energy of 7 mJ was used. The ion trap configuration and the whole experimental setup is shown in figure 8.4. The center electrodes, on which the rf potential is applied are 3 cm long, while the endcaps have a length of 1.5 cm and 1 cm. The middle electrodes have a DC offset of 75 V, which defines the kinetic energy of the ions when they were extracted in direction of the channeltron. The best shoot out efficiency was reached for DC potentials as illustrated in figure. 8.4.

#### 8.2.2 Measuring scheme

In figure 8.4 the trigger scheme is illustrated. The power of the Nd:YAG laser is adjusted by changing the delay between flash lamp and internal Q-switch. To avoid continuous creation of new ions in the trapping region a coincidence unit between delay and gate generator (DGG) 1 and the delayed flash lamp trigger defines the number of laser pulses which are shot into the trap for non-resonant ionization. The output signal of DGG 1 is the start trigger for DGG 2. After a delay, which corresponds to the trapping time the DC trap potentials are changed for a short time, which is defined by the gate length to release the ions that have been trapped.

For a better suppression of the background a third DGG is used, which opens a gate with a length of 200  $\mu$ s in time with the output signal of DGG 2. The channeltron signal is amplified, shaped in a "Timing Filter Amplifier" (TFA) and discriminated. The TTL/NIM signal is fed into a coincidence unit with the signal of DGG 3 and is read out with a counter card (PXI-system) for a specified number of cycles. This process is repeated, until the output signal of DGG 3 is interrupted before it starts DGG 1.



Figure 8.6: Trapping efficiency in dependence of various parameters: (A) RF-amplitude, (C) Time of ion creation, (D) Trapping time. (B) illustrates the probable saturation of the channeltron. Further information can be found in the text.

#### 8.2.3 Results

At first, the trap was tested with different rf amplitudes varying from 200 to 450 V<sub>pp</sub>, constant trapping (500 ms) and ion production times (1 s). The result is shown in figure 8.6 (A). Simulations indicate that the observed relation of increasing count rate with increasing rf potential, does not correspond to a higher trapping efficiency, but rather to a higher extraction efficiency. Since the higher rf amplitude is equal to a higher stability parameter, the ions are squeezed closer on the trap axis, which reduces ion losses due to collisions with the support structure of the trap during the extraction process. The small counting rates are probably a result of the impinging ion bunch which saturates the channeltron, i.e. not all ions are counted individually. All counting rates as shown in this section have been recorded in a time window of 200  $\mu$ s, while the ion bunch seems to have a duration of less than 100  $\mu$ s as shown in figure 8.6 (B), where the amplified channeltron signal (TFA) is plotted in comparison to the trigger that starts the ion release.

Another property that was tested is the storage capacity of the trap. For this purpose ions were produced for time intervals between 100 ms and 4 s, while the rf amplitude  $(300 V_{pp})$  and trapping time (500 ms) were kept constant. A strong increase of the counting rate is observed, which reaches a plateau after 1 s (figure 8.6 (C)). This behavior can be explained by the fact, that the trap is filled with a maximum number of ions. Further ions can not be captured due to space charge effects in the trap. But also here no absolute number of stored ions can be extracted, because of the saturation of the channeltron.

The most important property that was tested is the storage time of the ions. In this case the ions were stored for some ms up to 8 s while the rf amplitude (300  $V_{pp}$ ) and ion production time (1 s) were kept constant. The result of this measurement is shown in figure 8.6 (D). The mean storage time was approximately 1 s, which is rather short. But it can be supposed that the results are realistic, since the experimental conditions were unfavorable: The pressure inside the vacuum chamber was as high as  $10^{-6}$  mbar and the atom flux from the oven was assumed to be quite high. This leads to a large number of collisions between stored ions, residual gas and Li atoms, which dramatically reduce the storage time.

## 8.3 First trapping of $Be^+$ - optical

The last test, which was performed during this work is the optical detection of trapped ions. In this case Be atoms are produced by evaporation of a metallic Be wire and are ionized by electron impact ionization. The whole experimental setup is illustrated in figure 8.7 and will be discussed in this section.

#### 8.3.1 Ion source

In first off-line tests the Be ions are directly produced inside the trapping volume. Be is a brittle material, which has a very high melting point of approximately 1300°C and a high ionization potential of 9 eV. Evaporation and ionization of such a material is a non trivial issue. Heating in funnels as used in mass spectroscopy [GEP05], can not be used because Be diffuses into the Carbon surface and desorption from metallic Be foils is not suitable, because Be ions with a rather big energy spread are produced [MAU05, OKA98]. Another technique for the evaporation of Be is described in many publications [SCH01, KIN92]: A thin Be wire with a diameter of 50  $\mu$ m is wrapped several times around a 150  $\mu$ m tungsten wire. To increase the flexibility of the brittle Be wire, it was heated with a heat gun during the wrapping process. In general 10 cm of Be wire are wrapped around a 1 cm tungsten wire with a covering ratio of approximately 40-50 %. The tungsten wire is connected to electrodes in a closed stainless steel housing, which is mounted 5 cm above the ion trap. The housing is needed to reduce the contamination of the whole vacuum chamber with Be and has an aperture of  $1 \times 15 \text{ mm}^2$ , which is pointing to the trap.

In [SCH01] it is estimated that an heating temperature of 800 to  $900^{\circ}C$  is sufficient to achieve appropriate loading rates of some hundred ions per second, which means moderate heating currents of 1.5 to 2 A, which is in well agreement with [ROT07]. The evaporated Be atoms are overlapped with an electron beam inside the ion trap. The optimum electron energy for electron impact ionization is 50 eV for Be. In this experimental setup this energy has to be increased to 750 eV [ROT07], because the electrons have to pass the trap electrodes, which are driven at rather high voltages.



Figure 8.7: Experimental setup for optical detection of stored Be ions. The cooling laser is directed into the trap along the trap axis. A  $\lambda/2$  plate is used to adjust the polarisation to  $\pi$  polarisation in the y direction. Also the Be ion source, the applied trapping potentials and a technical drawing of the EMCCD device as well as the support structure is shown. The rf potential is only applied on the middle trap electrodes.



Figure 8.9: The left plot shows how the effective readout noise is reduced when the electron multiplying is increased. The right plot shows the quantum efficiency of the Andor:iXon.

Table 8.1: Derived objective properties of the used lens system.

Focal length $f$	75.32	mm
Opening $D$	32	mm
Image distance	861.24	$\mathbf{m}\mathbf{m}$
Resolution	1.3	$\mu { m m}$
Depth of focus	$\sim 20$	$\mu m$

#### 8.3.2 Optical detection system

The optical detection system that is needed for efficient detection of fluorescence photons was developed and constructed as well as the support construction at the university in Ulm and will be described in more detail in [EBL09, MAI06]. For a reduction of spherical and chromatic aberration a set of 4 lenses is used to image the fluorescence photons and was produced from "Sill Optics" and specially anti-reflection coated for 313 nm. The data sheet of this objective could be

Table 8.2: Calculation of the overall detection efficiency.

Solid angle	2.5	%
UV filter	64	%
Transmission viewport	95	%
Transmission objective	90	%
Quantum efficiency	10	%
Total efficiency	0.14	%



Figure 8.8: Simulation of the optical path of the used objectiv for a point like light source [EBL09]

used to derive its optical properties, which can be found in table 8.1. For illustration purposes figure 8.8 shows the simulated optical path of a point like light source like (e.g. an ion at rest) through such a lens system. The fluorescence light is imaged on an electron multiplier charged coupled device (EMCCD) from Andor (type: iXon). It has a CCD chip with  $128 \times 128$  pixels, which have a size of  $24 \times 24 \ \mu m^2$ . Thermally induced dark counts

are reduced by Peltier cooling of the CCD chip to -100°C. A further improvement of the signal to noise ratio by more than three orders of magnitude is achieved by the included electron multiplier, which is shown as well as the quantum efficiency<sup>3</sup> in figure 8.9.

Objective and EMCCD device are mounted on a L-shape support construction, which is connected to an x, y, z positioning table. This setup was necessary, because space for the experimental setup at ISOLTRAP is rare. The used setup allows the adjustment of the focal point along the trapping volume. Y and z direction are totally independent, while the x direction is unfortunately coupled to the y position, which is an uncomfortable property during the alignment. An illustration of the whole assembly can be found in figure 8.7.

The expected fluorescence rate can easily be calculated. The photons are scattered in a solid angle of  $4\pi$ , but the objective can image only  $\frac{1}{10}$  of  $\pi$ . Also the limited transmission of viewport and objective as well as the quantum efficiency of the EMCCD device decrease the recorded scattering rate as summarized in table 8.2. With the natural linewidth and the resulting scattering rate of 34 MHz for the  $2s_{1/2}(F = 1) \rightarrow 2p_{1/2}(F = 2)$ transition of <sup>7,9</sup>Be ions one can calculate the expected detection rate to be approximately 47 kHz. In [MAI06] it is shown that a crucial point of such a detection system is the depth of focus. With only some  $\mu$ m it is rather small, which is a result of the large solid angle that has to be imaged. When the focal plane is wrong by only 18  $\mu$ m the recorded fluorescence intensity is reduced by 50 %..

#### 8.3.3 Results

Unfortunately it was not possible to detect any fluorescence signal of trapped Be ions so far. Both transitions, the D1 and the D2 line were used for cooling. Avoiding of the population of dark states was tried by optical pumping on the  $2s_{1/2}(F = 2, m_F = 2) \rightarrow 2p_{3/2}(F = 3, m_F = 3)$  transition as well as by microwave induced mixing of the ground state hyperfine levels. There are several possible reasons, that may explain this failing of the optical detection:

- 1. The ionization process with the electron beam is not efficient enough to produce a large amount of Be<sup>+</sup> ions.
- 2. The power of the microwave, irradiated for mixing of the ground state hyperfine levels is too small.
- 3. The polarization optics which have been specially ordered for 313 nm show strange behavior and do not work properly.
- 4. The vacuum with 5  $10^{-8}$  mbar is possibly too bad and produced Be<sup>+</sup> ions are kicked out of the trap by collisions with residual gas.
- 5. Possible charging effects or problems with the electronics.

<sup>&</sup>lt;sup>3</sup>Wavelength sensitivity of the CCD device.

#### 8.3. FIRST TRAPPING OF BE+ - OPTICAL

Because it is difficult or even impossible to test those points separately, I suggest the test of the ion trap with an element that can be treated more easily. For the future test with Ba<sup>+</sup> are considered where additional buffer gas cooling can be applied.

# Chapter 9 Summary and Outlook

In this thesis two different approaches for the spectroscopy of radioactive Be<sup>+</sup> ions have been prepared. The goal was to measure the isotope shift of <sup>7,9,10,11</sup>Be<sup>+</sup> and the extraction of the nuclear charge radii. Spectroscopy of laser cooled Be<sup>+</sup> ions in a linear Paul trap yields high resolution measurements of optical transition frequencies, isotope shifts and hyperfine anomalies [GRO99]. The ion optics required for an efficient transport of ions from the ISOLTRAP RFQ cooler and buncher towards the Paul trap and the pulsed drift tube for the deceleration of fast ions were simulated, constructed and partially tested.

Two ion trap designs were developed and ion storage in the standard trap design with cylindrical rods was demonstated. However, due to limited availability of the laser system and problems with the ISOLTRAP off-line ion source, the RFQ and the laser system laser cooling and optical detection of the captured ions have not been realized within the time of this thesis.

The nuclear charge radii of the beryllium isotopes  $^{7,10}$ Be and the one neutron halo nucleus  $^{11}$ Be were determined for the first time by measuring the isotope shift in the  $2s_{1/2} \rightarrow 2p_{1/2}$  transition of Be<sup>+</sup> ions in a fast ion beam. The extraction of the field shift contribution and therewith of the nuclear charge radius was possible by combining these measurements with high accuracy calculations of the mass shift contribution [YAN08, PUC08]. For this purpose a novel improvement for collinear spectroscopy on light elements (Z<10) has been developed at the Institute for Nuclear Chemistry of the Johannes Gutenberg-University in Mainz and was finally used at the COLLAPS experiment at ISOLDE/ CERN. The combination of simultaneous collinear and anti-collinear laser spectroscopy with the absolute frequency determination by a frequency comb made it possible to resolve the limitations of collinear laser spectroscopy, which are mainly introduced by the uncertainty of the acceleration voltage. With the described setup it is now possible to measure absolute transition frequencies on fast ions with a relative uncertainty of  $10^{-9}$  and isotope shifts with a relative uncertainty of a few times  $10^{-5}$ .

A first beam time at the COLLAPS experiment at ISOLDE/ CERN, Geneva, in April 2008 was used for the careful investigation of possible systematic error sources. Almost no systematic influences of the experimental setup could be found in the data analysis. Only a possible misalignment seems to contribute systematically with approximately 500 kHz to the total uncertainty. The isotope shifts between  $^{7,9,10,11}$ Be<sup>+</sup> were measured in June

2008. The laser ion source RILIS provided a yield of approximately  $10^{6}$  <sup>11</sup>Be ions/second which was sufficiently high for collinear laser spectroscopy.

The derived nuclear charge radii decrease continuously from <sup>7</sup>Be to <sup>10</sup>Be and increases again for <sup>11</sup>Be by approximately 4.5 % [NOE09]. The results have been compared with theoretical predictions of different nuclear models. All models clearly exhibit a similar trend as the obtained experimental values. Particularly good agreement was found for Greens function Monte Carlo calculations and the Fermionic Molecular Dynamics model.

While the ion trap spectroscopy will potentially provide higher accuracy for isotope shift measurements, they will not allow measurements of very short lived isotopes like <sup>12</sup>Be  $(t_{1/2} = 20 \text{ ms})$  and <sup>14</sup>Be  $(t_{1/2} = 4 \text{ ms})$ . With collinear laser spectroscopy these isotopes could be investigated provided that sufficiently high ion yields are available. At ISOLDE the production yields for <sup>12,14</sup>Be are very low and reach only 1500 and 4 ions/second, respectively, which is too low for the described type of collinear laser spectroscopy. Since the ISCOOL cooler and buncher at ISOLDE is operational, it is possible to improve the signal to noise ratio by a factor of  $10^4$ . With this technique it might be possible to determine the nuclear charge radius of <sup>12</sup>Be<sup>+</sup> in the near future. Tests, if ISCOOL is an appropriate tool for cooling and bunching of Be<sup>+</sup>, are in preparation. Nevertheless, the isotope shift of <sup>14</sup>Be will not be feasible at ISOLDE if the yield is not increased at least two orders of magnitude. For this isotope the applied production method is not suitable and isotope in-flight production seems to be more appropriated. However, the produced ions must be stopped in a gas cell and extracted to provide an ion beam with sufficiently high quality. Wether this will be possible must be tested before an isotope shift measurement seems to be realistic. Such measurements may be performed in the future at the LASPEC setup at the low-energy beam line of the FAIR super-target recoil separator (S-FRS).

# Appendix A Trap type 2 - Circuit board



Figure A.1: The layout of DC and RF electrodes is shown as well as the support structure. Further information can be found in the text.

The circuit board type trap was manufactured at the company micro pcb ag<sup>1</sup> in Switzerland. The used wafer has a thickness of only 0.4 mm and is made of epoxy resin. It is coated with a copper layer that has a thickness of some  $\mu$ m that can be additionally coated with gold to achieve best trapping potentials [SCH09]. In principle any desirable electrode configuration can be produced. The trap geometry that was constructed during this thesis is illustrated in figure A.1.

Here, only two segments are used for axial or radial ion confinement, respectively. The wafers for axial ion confinement are structured into 18 segments with a length of 2 mm and 6 segments with a length of 1 mm that are separated by 0.2 mm. All circuit paths lead to one side of the trap, where DC and RF potentials can be applied. The wafers for radial ion confinement are structured the same way to avoid possible fringe fields, but they are all bypassed approximately 1 cm above the trap axis. The surface that is not used for the trapping potential is also coated with copper and connected to ground potential

<sup>&</sup>lt;sup>1</sup>www.micro-pcb.ch



Figure A.2: Support structure for the circuit boards. Further information can be found in the text.

to avoid charging effects during the experiments. The tapering on the left side of the circuit boards and at the transit between big and small electrode segments, as illustrated in figure A.1, were thought to increase the capturing and transfer efficiency.

These wafers are connected to a support structure that is shown in figure A.2. It is made of titan and has an extreme rigid structure to guarantee best alignment accuracy. All wafers are connected with four screws and can be easily exchanged, if another wafer layout has to be used.

# Appendix B

# Circuit diagrams of the rf-amplifier

#### Extract of [LEN07]:

#### Transistordifferenzverstärker

Das Eingangssignal an X8 steuert über den Hochfrequenztransformator TR3 (minicircuit T2.5-6T) die Basen der Transistoren T3 (NTE2633) und T4 (NTE2633) im Gegentakt. R13 und R15 sichern eine Abschlußimpedanz von 50 $\Omega$ . Die Emitter der Transistoren T3 und T4 sind über R11 (47 $\Omega$ ) und R12 (47 $\Omega$ ) mit dem Kollektor von Transistor Q1 (BD139-10) verbunden, der als Stromquelle arbeitet. Der Konstantstrom wird mit dem Trimmer TR4 eingestellt und ermöglicht ein gleichseitiges Verschieben der Basisspannungen der Kaskadenransistoren T1 (NTE2506) und T2 (NTE2506). T1 arbeitet als Stromquelle, dessen Kollektorstrom bestimmt wird von seiner Basisspannung und dem Emitter-Widerstand R9 (10 $\Omega$ ). Das selbe gilt für T2 und R10. [...]

#### Röhrenleistungsstufe mit Resonanzkreis

Transistor T1 bildet zusammen mit der Röhre Rö1 (C1108, QB 3/300) eine Kaskadenverstärkerstufe. Das Steuergitter g1 der Röhre Rö1 liegt dabei auf Massepotential. Um Selbsterregung bei ca. 150MHz zu unterdrücken, sind die Steuergitter über Parallelschaltungen von  $10k\Omega$  und  $100\mu$ H mit Masse verbunden. Die Gitter-Kathodenspannung stellt sich selbständig so ein, daß durch Röhre und Transistor der gleiche Strom fließt, der weitgehend unabhängig von der Anodenspannung ist. [...] C21 (10nF) und C22 überbrücken die Heizfäden kapazitiv mit ca.  $1\Omega$ . Die Schirmgitter g2 liegen auf 350V und sind über C15 (10nF) und C16 nach Masse geblockt. Der Anodenschwingkreis besteht aus der Luftspule L4 (0.9uH) dem Doppeldrehkondensator C14 (2x23..220pF), den Kabelkapazitäten und der Ion-Guide-Kapazität. W[...]

#### Bias-Spannungs-Netzwerk mit Monitoranschlüsse

C11 bis C13 (4.7nF) blocken die Anodenspannung zu den Ausgängen ab. Diese Kondensatoren sind bis 4kV spannungsfest. Über die kreuzgewickelten Drosseln L1 (5mH) und L2 (5mH) ist es möglich die Ausgänge A und B unabhängig voneinander auf Gleichspannungspotentiale zu setzen. Dabei ist zu beachten, daß die Spannungsdifferenz zur Anodenspitzenspannung von +1500V 4000V nicht überschritten wird. Als Hilfe zum einstellen des Resonanzmaximums und der Symmetrie der Ausgangsspannungen dienen die kapazitiven Spannungsteiler C2/C3 (3.9pF/330pF) und C6/C7. Zusammen mit ausgedienten Prüfspitzen für Oszillographen bilden diese 1000/1-Spannungsteiler zum Anschluß an Messgeräten mit 1M $\Omega$ /10pF-Eingängen. [...]



Figure B.1: Electronic circuit of the used RF amplifier. Developed in the electronics workshop of the institute of physics in Mainz by Heinz Lenk.



Figure B.2: Electronic circuit of the needed power supply for the RF tube amplifier. Developed in the electronics workshop of the institute of physics in Mainz by Heinz Lenk.
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