An ion trap for laser spectroscopy on lithium ions

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Abstract

An experimental setup for the spectroscopy of metastable singly-charged lithium ions, confined in a linear Paul trap, and sympathetically cooled by laser cooled magnesium ions has been built. It consists of an ultra-high vacuum chamber reaching pressures down to $5 \times 10^{-11}$ mbar, an ion trap with associated electronics, as well as an imaging system for optical detection and laser system. The trap has been taken into operation and fully characterized by loading and laser cooling of magnesium ions generated via photoionization and electron-impact ionization.

A possible approach towards spectroscopy on lithium II as well as its impact on two-electron bound-state QED is discussed.
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Finally, I thank my parents and family. You gave me a lot of freedom
and always trusted in me believing that I have a right to make my own mistakes. Thank you.
1

Introduction

Our deepening understanding of Nature and technical progress has been always pushing the frontiers of our perception of "precision". Investigation of a pendulum and discovery of its isochronism allowed Galileo to establish kinetic equations of motion. Based on this research Christiaan Huygens developed a pendulum clock pushing the accuracy of time measuring from 15 minutes per day to about 10 seconds per day. 270 years later quartz clocks redefined "precise" to 10 seconds per year. In recent years this "quest for precision" accelerated and the frontiers of the unknown have been pushed further and further. For physics it resulted in new, revolutionary discoveries that changed the face of our world. In the early years of quantum mechanics spectroscopic data led Bohr to his model of the hydrogen atom. Improved measurements revealed deviations from the description Bohr proposed so that this model was replaced by the Schrödinger equation later modified to incorporate the electron spin in order to account for the fine structure. This development led to a fully relativistic description of the hydrogen atom by the Dirac equation. 20 years later more accurate measurements of hydrogen spectra revealed another deviation, the Lamb shift, which resulted in the birth of quantum electrodynamics (QED). The history of science proved that when we persistently look for details with better tools and methods it is always possible that the next decimal number we get from the measurements can reveal some new knowledge, previously hidden behind the noise of the older data, leading to new and quite often revolutionary discoveries. For years it has been hydrogen, that drove the progress of our understanding of the word "precise". In recent years, however, attention was extended to more complicated systems with heavier nuclei, which allow studying effects
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not present in hydrogen. Despite their complexity enormous improvements in the accuracy of measurements made them competitive for some applications, like testing of bound-state QED.

To a large extent this progress is possible owing to the development of ion trap techniques [1, 2] which provide an unprecedented level of control over the system under investigation. Ion traps make it possible to study "substitutes" of hydrogen and helium: He$^+$[3] and Li$^+$ (this work), respectively, which can open new possibilities of testing quantum electrodynamics. Considered as the best confirmed theory ever, in fact, QED in its bound-state version has not been that precisely tested as for free particles.

Problems, that arose for helium from discrepancies between theory and experiments show, that QED calculations are not fully understood. This work is a step towards trapping lithium ions, which suppose to help solving the problems encountered in the structure of He and other two-electron atoms.

The thesis is organized as follows. Chapter 2 contains an introduction to two-electron atoms and presents problems arising from discrepancies between theory and measurements for helium. The basic properties of lithium ions are introduced followed by a discussion of advantages and disadvantages of Li$^+$ for precision spectroscopy. Chapter 3 presents the theory of ion traps, laser cooling and the use of magnesium ions as a coolant in Li$^+$ measurements, the backbones of the planned measurement scheme. Chapter 4 describes an operational ion trap setup, which was built in the framework of this thesis. In Chapter 5 initial results of an experiment with trapped ions are presented. Finally Chapter 6 summarizes the progress towards trapping of lithium ions and discusses further steps that should be taken towards spectroscopy on Li$^+$. 
2

Spectroscopy on simple atomic systems

2.1 One-electron bound-state QED

Hydrogen spectroscopy has played an important role in the development of physics. As an only two-body system hydrogen was relatively easily accessible to calculations. Continuous improvements in the measurement, especially, of its $1S - 2S$ transition have led to the development of modern quantum mechanics, first non-relativistically with the Schrödinger equation, then relativistically through the Dirac equation, and finally to quantum electrodynamics (QED), which takes the vacuum into account. The quest for the best possible measurements of the hydrogen’s spectra has been vivid for more than a century now, creating not only great scientific results but also contributing to the development of modern technologies like laser frequency stabilization techniques or frequency combs. Hydrogen measurements are limited now by nuclear effects, especially by the proton size, which is the biggest source of uncertainty, which is not expected to be reduced in the future.

2.2 Two-electron QED

The investigation of the next simplest atom, the helium atom, in the early days of quantum mechanics has also played a significant role. Hylleraas’ treatment [4] of the helium atom has proven the validity of the Schrödinger equation for atoms more complex than hydrogen. Later high-precision tests in two-electron systems were for a long
2. SPECTROSCOPY ON SIMPLE ATOMIC SYSTEMS

time hampered by the fact that the non-relativistic two-electron atom, which is a three-body system, is not exactly solvable. This prevented the calculation of non-relativistic energies to accuracies were higher order correction played a role. Additionally it led to a lack of suitable wave functions for perturbation calculations of relativistic and QED corrections. This principle problem has been solved in the 90s by Drake and coworkers (for references see e.g. [5]).
The availability of essentially exact non-relativistic wave functions for the entire spectrum of helium and helium-like ions now allows for precise theoretical calculations of several interesting features of these systems:

- He QED allows to study correlation effects that are absent in hydrogen but are essential for all many-electron atoms. He is thus a prototype for complex atoms,
- The fine structure of the triplet P levels allows for the determination of an atomic physics value of the fine structure constant,
- Isotope shift measurements can be exploited to extract information about the nuclei.

**Theory of two-electron atoms**

The description here follows that of [6]. It starts with the non-relativistic two-electron Hamiltonian for infinite nuclear mass:

\[
H = \frac{p_1^2}{2m} + \frac{p_2^2}{2m} - \frac{Ze^2}{4\pi\epsilon_0 r_1} - \frac{Ze^2}{4\pi\epsilon_0 r_2} + \frac{e^2}{4\pi\epsilon_0 r_{12}},
\]

(2.1)

with \(r_1, r_2\) and \(r_{12}\) defined in Fig. 2.1 Due to the last (correlation) term, the Schroedinger Equation

\[
H\Psi(r_1, r_2) = E\Psi(r_1, r_2)
\]

(2.2)

is not separable and, as a consequence, not exactly solvable. The most successful approximation method is based on variation calculus, which searches for solutions that fulfill the variational condition

\[
\delta \int \Psi^*(H - E)\Psi d\tau = 0.
\]

(2.3)
2.2 Two-electron QED

Following an ansatz of Hylleraas \[4\], the wave functions can be expanded in terms of states \(Y_{l_1,l_2,L}^{M}(r_1,r_2)\) of total angular momentum \(L\) that are constructed from spherical harmonics:

\[
\Psi(r_1,r_2) = \sum_{i,j,k} a_{ijk} r_1^i r_2^j r_{12}^k e^{-\alpha r_1 - \beta r_2} Y_{l_1,l_2,L}^{M}(r_1,r_2). \tag{2.4}
\]

\(a_{ijk}\) are the variational parameters, and \(\alpha\) and \(\beta\) scale factors to optimize the energy. It can be shown that this basis set is complete when all powers \(i, j, k\) up to infinity are included. The calculation is then done successively by including powers with \(i+j+k < \Omega\) for increasing integer numbers \(\Omega\) until the result converges. The Hylleraas basis set turned out to be very efficient for the description of correlation as only few powers were needed to recover the correlation energy represented by the last term of the Hamiltonian (Eq. 2.1). However a large number of powers needed to be taken into account to achieve a numerical accuracy of better than \(10^{-9}\). Moreover the accuracy quickly deteriorated for large quantum numbers \(n\). As a consequence, the approximate energy solutions and the corresponding wave functions of the non-relativistic two-electron atom were not accurate enough to deduce high precision tests of QED from comparison with experimental data. The solution to this longstanding problem was developed by Drake.
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and co-workers by doubling the Hylleraas basis set:

\[
\Psi(r_1, r_2) = \sum_{i,j,k} a_{ijk}^{A} r_1^{i} r_2^{j} Y_{l_1,l_2,L}^{M}(r_1, r_2) + a_{ijk}^{B} r_1^{i} r_2^{j} Y_{l_1,l_2,L}^{M}(r_1, r_2)
\]

(2.5)

with independent variational parameters \(a_{ijk}^{A}, a_{ijk}^{B}\), and scale factors \(\alpha_{A,B}, \beta_{A,B}\), respectively. The greater flexibility allows for two different scales, which describe separately the strongly correlated part of the wave function close to the nucleus, and the long range part of the wave function, where correlation plays a minor role. This method not only further improved the accuracy of the low-lying states but also allowed a better description of the high-lying Rydberg states. The resulting energies of the entire spectrum converge much more rapidly than before with remaining variations below 3 parts in \(10^{22}\) so that the non-relativistic two-electron atom is considered to be solved. The essentially exact solution of the non-relativistic problem provides the gross structure of the spectrum; all other relativistic, QED and nuclear effects are significantly smaller and can be calculated perturbatively by expanding the corresponding Hamiltonians in terms of the fine structure constant \(Z\alpha\) and the ratio of the electron’s reduced mass and the nuclear mass \(\mu/M\). The following table comprises the expansion order of various contributions to the energies:

<table>
<thead>
<tr>
<th>Contribution</th>
<th>Magnitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nonrelativistic energy</td>
<td>(Z^2)Ry</td>
</tr>
<tr>
<td>Mass polarization</td>
<td>(Z^2\mu/M)Ry</td>
</tr>
<tr>
<td>Second-order mass polarization</td>
<td>(Z^2(\mu/M)^2)Ry</td>
</tr>
<tr>
<td>Relativistic corrections</td>
<td>(Z^4\alpha^2)Ry</td>
</tr>
<tr>
<td>Relativistic recoil</td>
<td>(Z^4\alpha^2\mu/M)Ry</td>
</tr>
<tr>
<td>Anomalous magnetic moment</td>
<td>(Z^4\alpha^3)Ry</td>
</tr>
<tr>
<td>Hyperfine structure</td>
<td>(Z^3g_1\mu_0^2)Ry</td>
</tr>
<tr>
<td>Lamb shift</td>
<td>(Z^4\alpha^3\ln\alpha)Ry + ...</td>
</tr>
<tr>
<td>Radiative recoil</td>
<td>(Z^4\alpha^3\ln\alpha\mu/M)Ry</td>
</tr>
<tr>
<td>Higher-order QED</td>
<td>(Z^5\alpha^4)Ry + ...</td>
</tr>
</tbody>
</table>

Table 2.1: Contributions to the energy and their magnitude
Recoil corrections

All terms of the α expansion are first calculated for infinite nuclear mass. Subsequently recoil corrections due to finite nuclear mass are added perturbatively by successive orders of μ/M until their contributions are negligible on the level of desired accuracy. The recoil correction to the non-relativistic energies are sufficiently taken into account by the first and second order mass polarization term.

Relativistic corrections

The relativistic corrections to order α² can be deduced from an approximate relativistic wave equation of the two-electron atom, the Breit equation

\[ H_{DCB} \Psi(r_1, r_2) = E \Psi(r_1, r_2). \] (2.6)

The Dirac-Coulomb-Breit Hamiltonian \( H_{DCB} \) is given as

\[ H_{DCB} = H_D^{(1)} + H_D^{(2)} + \frac{1}{4\pi\varepsilon_0 r_{12}} + B_{12}, \] (2.7)

where the non-relativistic one-electron Hamiltonians are replaced by the corresponding Dirac-Hamiltonians

\[ H_D^{(i)} = c\alpha_i \cdot p_i + \beta_i mc^2 - \frac{Ze^2}{4\pi\varepsilon_0 r_i} \] (2.8)

with the Dirac matrices \( \alpha \) and \( \beta \), and the instantaneous electron-electron interaction of Eq. 2.1 is modified by the Breit interaction

\[ B_{12} = \frac{e^2}{8\pi\varepsilon_0 r_{12}}[\alpha_1 \cdot \alpha_2 - \frac{(\alpha_1 \cdot r_{12})(\alpha_2 \cdot r_{12})}{r_{12}^2}], \] (2.9)

which takes retardation and magnetic interaction to order \( \alpha^2 \) into account. Although not strictly Lorentz-invariant, the Breit equation has been shown sufficiently accurate to include relativistic corrections to order \( \alpha^2 \).

Within the Pauli approximation, the Dirac-Coulomb-Breit Hamiltonian can be written as a sum of various energy contributions:

\[ H_{DCB} = H_0 + H_1 + H_2 + H_3 + H_4 + H_5. \] (2.10)

Here, \( H_0 = \frac{p_1^2}{2m} + \frac{p_2^2}{2m} - \frac{Ze^2}{4\pi\varepsilon_0 r_1} - \frac{Ze^2}{4\pi\varepsilon_0 r_2} \) is the non-relativistic Hamiltonian and \( H_1 \ldots H_5 \) are relativistic corrections of order \( \alpha^2 \) with the following physical significance identified in [7]:

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- $H_1$: relativistic kinetic energy correction
- $H_2$: orbit-orbit interaction due to magnetic interaction and retardation
- $H_3$: spin-orbit coupling
- $H_4$: the Darwin contact term
- $H_5$: spin-spin interaction

The latter three terms are spin-dependent and are thus relevant for the fine structure. Additionally all these terms are derived under the assumption of infinite nuclear mass and thus have to be corrected for the finite nuclear mass yielding correction terms of order $\alpha^2 \mu/M$.

All these corrections are known to much higher accuracy than the lowest order OED corrections which thus become significant.

**QED corrections**

The QED corrections two the two-electron atom are usually classified as

$$E_{QED} = E_{L,1}^{(3)} + E_{L,2}^{(3)} + O(\alpha^4 Ry).$$

The index '3' indicates corrections of order $\alpha^3 Ry$, the lowest-order QED correction which is commonly defined as the Lamb shift. For each order of $\alpha$ there are two contributions: $E_{L,1}$ are corrections similar to those of the hydrogen atom which arise from the electron-nucleus interactions. Due to the $Z^4$-scaling they are however significantly larger than in H. $E_{L,2}$ describes terms from the electron-electron interaction, which are absent in hydrogen, but play a significant role not only in He-like ions but for many-electron atoms in general. To orders $\alpha^3 Ry$ and $\alpha^4 Ry$ these terms and the lowest-order recoil corrections of order $\alpha^3 Ry\mu/M$ are known for all states of helium [8]. The estimated uncertainty due to higher-order terms are of the order of 1 MHz for the $n = 2S$ states and 400 kHz for the $n = 2P$ states. There is moderate agreement between theory and experiment on this level except for the $^1P_1$ state.
2.3 $^3P_{0,1,2}$ fine structure in helium

While the investigation of the hydrogen atom has resulted in a very precise value of the Rydberg constant, it is not well suited to derive a precise atomic physics value of the fine structure constant. This is because the $P$ levels which exhibit fine structure are rather short-lived and thus suffer from a large natural linewidth of about 120 MHz. In helium, on the other hand, the triplet $P$ states have a linewidth of 2 MHz, almost two orders of magnitude smaller and thus allowing for precise measurements. Additionally the fine structure measurement in He is not obscured by hyperfine structure as the $^4\text{He}$ nucleus has spin $I = 0$. In order to derive $\alpha$ to an accuracy compatible with other experiments, the experimental and theoretical values of the fine structure need to be compared on the level of about 1 kHz. Fine structure is a purely relativistic effect that arises from the spin-dependent terms of the Breit Hamiltonian (or Dirac Hamiltonian in the case of one-electron atoms), which scales as $\alpha^2$. The procedure to extract $\alpha$ is to subtract all significant higher-order QED corrections from the measured value and then adjust $\alpha$ to match it with the leading $\alpha^2$ theoretical value. To this end, the relevant QED-corrections must be known to an accuracy better than the anticipated one of 1 kHz. Drake [9] and Pachucki [10] have independently put tremendous work into the calculation of all higher-order QED corrections up to the order $\alpha^5\text{Ry}$ and recoil corrections up to order $\alpha^5\text{Ry}\mu/M$ to the spin-dependent terms of the Breit interaction. The following table shows their results and a comparison with recent experiments. For the calculation of the fine structure intervals the CODATA value of $\alpha$ was used.

Until recently the theoretical uncertainty due to neglected higher-order QED terms was estimated to be below 200Hz. However there is a strong disagreement between theory and experiment that cannot be reasonably accounted for by an adjustment of $\alpha$. Recently Pachucki and Yerokhin [18] reexamined the calculations and performed a new estimation on the size of the neglected terms. Nevertheless a disagreement prevails and their calculations agree only for the small interval $\nu_{12}$, whereas the large interval $\nu_{01}$ still shows discrepancies about three standard deviations from the experiments. The measurements of the three $^2P_j$ fine structure intervals $\nu_{12}$, $\nu_{01}$ by Zelevinsky [15] showed inconsistencies in the two-electron QED since only $\nu_{12}$ is correctly described by the theory.
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<table>
<thead>
<tr>
<th>Experiment</th>
<th>$\nu_{01}$</th>
<th>$\nu_{12}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Minardi et al. [11]</td>
<td>29 616 949.7(2.0)</td>
<td>-</td>
</tr>
<tr>
<td>Castillega et al. [12]</td>
<td>-</td>
<td>2 291 175.9(1.0)</td>
</tr>
<tr>
<td>Storry et al. [13]</td>
<td>-</td>
<td>2 291 174.0(1.4)</td>
</tr>
<tr>
<td>George et al. [14]</td>
<td>29 616 950.9(9)</td>
<td>-</td>
</tr>
<tr>
<td>Zelevinsky et al. [15]</td>
<td>29 616 951.66(70)</td>
<td>2 291 175.59(51)</td>
</tr>
<tr>
<td>Giusfredi et al. [16]</td>
<td>29 616 952.7(1.0)</td>
<td>2 291 168.0(11.0)</td>
</tr>
<tr>
<td>Borbely et al. [17]</td>
<td>-</td>
<td>2 291 177.53(35)</td>
</tr>
</tbody>
</table>

**Table 2.2**: Comparison of theory and experiment for the fine structure splittings of helium $^2P_j$. Units are in kHz.

2.4 Lithium II

The above presented features of two-electron systems were mainly exploited in measurements of helium atoms and highly ionized atoms. To bridge the gap between these measurements few groups focused on the lightest, apart from helium, two-electron system: lithium II. Early works (e.g. [19]) showed, that lithium II can be indeed an attractive candidate for measurements, nevertheless it has never been as widely studied experimentally as helium. One of the reasons is that the $P$ states are accessible only in metastable ions, which complicates measurements due to their decay to the ground state. Recent results on helium, however, revealed some discrepancies between theory and experiments and to our knowledge they seem not to be easily overcome. On the other hand, the tremendous improvements in the theory of two-electron atoms makes Li$^+$ an interesting system to study. Due to the scaling with high powers of $Z$ (Table 2.2), the relevant QED terms currently under investigation in He, could be more easily accessible in Li$^+$. This way additional experimental data set on the two-electron system from Li$^+$ could help solving the mentioned problems in helium.

To date, lithium II measurements were mainly performed on ion beams generated from atomic beams. Only one approach, in the early 80’s, employed ion trap techniques,
however it was limited to a lifetime measurement of the metastable $2^3S_1$ state \[20\]. Lithium ions are charged, therefore with enormous progress in ion trap technology it was proposed to perform spectroscopic measurements of trapped ions under ultra high vacuum conditions \[21\]. Ion traps provide a very well controllable environment, in which Li$^+$ ions can be laser cooled on the cycling transition $2^3S_1 (F = 5/2) \rightarrow 2^3P_2 (F = 7/2)$ at 548 nm in order to reduce systematic effects like Doppler shifts and time-of-flight effects. Also sympathetic cooling can be applied, which in fact seems to be even more advantageous than direct cooling (see next Chapter). The natural lifetime of the $P$ states is two times shorter in Li$^+$ than in He so we assume, that the line center can be determined about two times worse than in He \[15\], with an accuracy down to 2 kHz. Larger fine-structure splittings ($\nu_{10} = 155.7$ GHz and $\nu_{12} = 62.7$ GHz) than in helium may lead to higher relative accuracy. It is expected, that with the measurement of the fine structure on the 2 kHz level it might be possible to determine $\alpha$ to 6 ppb if sufficient theoretical calculation will be available. To this end, however, so far unexplored terms of the order $\alpha^5 \text{Ry}$ need to be calculated. The anticipated result would be amongst one of the most precise measurements of the fine structure constant. However, not only accuracy is important. For testing QED it is advantageous to have $\alpha$ derived from as many systems as possible and this is in fact the main goal of the planned work. Also, the QED corrections scale at least as $Z^4$ (see Table 2.1) so it would be possible to verify if all neglected terms in helium theory were estimated correctly. The fine structure in Li$^+$ is predominantly due to the spin-spin term \[19\]. This fact means that the results in Li$^+$ can provide a more sensitive test for the correctness of the two-electron, spin-spin Hamiltonian than those in He, where spin-orbit and spin-spin terms contribute about equally.

**Hyperfine structure**

Although lithium II seems to be a very advantageous system for testing QED it has features that can make measurements much more complicated than in case of helium. The main difficulty arises from the presence of hyperfine structure. Both stable lithium isotopes $^6$Li and $^7$Li have nuclear spin $I = 1$ and $I = 3/2$, respectively thus to extract the fine structure the hyperfine structure shifts have to be derived from the measurements of the hyperfine structure splittings. To achieve this on the sufficient level of accuracy, additional input from nuclear theory may be necessary. On the other hand
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measurements of HFS in Li$^+$ when confronted with the calculations, can advance our understanding of nuclear structure effects in the HFS. Figure 2.2 summarizes the hyperfine energy structure of both $^6$Li$^+$ and $^7$Li$^+$.

Isotope shifts and nuclear charge radii

Measurements of isotope shifts of optical transitions and extracting from them the effect of finite nuclear size enables the accurate determinations of the charge radii of light nuclei. Atomic structure measurements provide a critical test of the effective low-energy nucleon-nucleon potential used to determine a theoretical value for the nuclear-charge radius [23] and other properties of nuclear matter. The finite nuclear volume produces a volume shift in the energy of each atomic state, given in lowest order by [24]

$$E_{\text{nuc}} = \frac{2\pi}{3} Z e^2 r^2 \langle \sum \delta(\vec{r}_i) \rangle$$

(2.12)

where $r$ is the mean square nuclear charge radius, $Z$ is the number of protons and $e$ is the proton charge. The summation term equals the expectation value of the electron density at the nucleus where $i$ is summed over all electrons of the atom. The difference in mean square nuclear charge radius $\Delta r^2$ between two isotopes such as $^6$Li$^+$ and $^7$Li$^+$ defined by

$$\Delta r^2 = r^2(^6\text{Li}^+) - r^2(^7\text{Li}^+)$$

(2.13)

could be also interesting as a reference for measurements of unstable isotopes $^8$Li and $^9$Li as well as the halo nucleus of $^{11}$Li [25].

The table comprises isotope shifts between $^6$Li$^+$ and $^7$Li$^+$ and $\Delta r^2$:

<table>
<thead>
<tr>
<th>Transition</th>
<th>Isotope shift [MHz]</th>
<th>$\Delta r^2$ [fm$^2$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li$^+$ $^2^3S_1 \rightarrow 2^3P_0$</td>
<td>34 747.73± 0.55</td>
<td>0.78± 0.06</td>
</tr>
<tr>
<td>Li$^+$ $^2^3S_1 \rightarrow 2^3P_1$</td>
<td>34 747.46± 0.67</td>
<td>0.78± 0.07</td>
</tr>
<tr>
<td>Li$^+$ $^2^3S_1 \rightarrow 2^3P_2$</td>
<td>34 748.91± 0.62</td>
<td>0.64± 0.06</td>
</tr>
</tbody>
</table>

Table 2.3: Measured isotope shifts and determination of the difference in mean square nuclear charge radius, after [26]
Figure 2.2: Energy level diagram of the of the $^6\text{Li}^+$ and $^7\text{Li}^+$ ions. - Fine- and hyperfine splittings given in GHz. Diagram based on [22]
2.4.1 Measurement scheme

Determination of the hyperfine structure splittings is not a straightforward task, therefore special attention has to be paid to the measurement of the complete HFS multiplet. Fig. 2.3 shows a schematic of the planned measurement scheme. At least two lasers are required. The spectroscopy (s) laser can be relatively weak (few mW), with narrow linewidth. The cooling (c) and re-pumping (r1, r2) beams come from a dye laser with an output of several hundred mW. High power is necessary because of losses in AOMs and EOMs. The AOMs and EOMs are used to shift the laser frequency to adjust it to the transitions of interest. The method used in the scheme is based on shelving [27, 28]. To fix the attention lets consider the measurement of the

\[
\begin{align*}
J & \quad F \\
0 & \quad 3/2 \\
2 & \quad 7/2 \\
1 & \quad 5/2 \\
1/2 & \\
3/2 & \\
5/2 & \\
7/2 & \\
\end{align*}
\]

Figure 2.3: Spectroscopy scheme for \(^7\)Li\(^+\) - proposed measurement scheme of (a) \(F = 3/2, 5/2, 7/2\) splittings (b) \(F = 1/2\) splittings. (bottom) laser system with a dye laser to generate the beams for the cycling and \(\Lambda\) transition respectively, as well as the re-pumping light. Using electro- and acoustooptic modulators three beams can be generated from one laser. The spectroscopy beams are generated by a diode laser.
2.4 Lithium II

$^{2}S_{2}$ ($F = 3/2 - F' = 5/2$) splitting. Laser $c$ drives a cycling transition scattering photons that are detected. The presence of fluorescence indicates that the observed ion is in the $F = 5/2$ state of the $^{2}S_{1}$ ground level. The spectroscopy laser $s$ is tuned to the $^{2}S_{1}$ ($F = 5/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 5/2$) transition. The excitation by $s$ is followed by the decay either back to the $^{2}S_{1}$ ($F = 5/2$), to the $^{2}S_{1}$ ($F = 3/2$), or to the singlet $^{1}S_{0}$ ion ground state. For the two latter cases a fluorescence drop due to the laser $c$ will be observed since photons are not scattered any more. The decay to the singlet ground state is permanent while to the $^{2}S_{1}$ ($F = 3/2$) can be re-pumped with the $r_1$ laser bringing the fluorescence back to the previous level. Only a successful sequence: fluorescence on-off-on is treated as an excitation event of the spectroscopy laser. The sequence is repeated many times by applying the spectroscopy laser at frequency $\nu_1$ for a well-defined time window. From the number of excitation events, an excitation probability at frequency $\nu_1$ is derived. The frequency is than changed to $\nu_2$ and the sequence starts again. As a result a histogram of the number of events versus $\nu_i$ is obtained. The closer the spectroscopy laser is to resonance with the transition the more events should be registered. Fitting a line shape model to the obtained diagram gives a center frequency of the transition. The laser $s$ is then tuned to the other transition $^{2}S_{1}$ ($F = 5/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 3/2$) and the procedure is repeated with the difference, that two re-pumping lasers $r_1$ and $r_2$ have to be used due to possible decay of the $^{2}P_{2}$ ($F = 3/2$) state to $^{2}S_{1}$ ($F = 5/2, 3/2, 1/2$) states. Subtracting the $^{2}S_{1}$ ($F = 5/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 3/2$) frequency from the $^{2}S_{1}$ ($F = 5/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 5/2$) frequency gives the $^{2}P_{2}$ ($F = 3/2 - F' = 5/2$) splitting. This method seems to be rather advantageous, because laser light can be controlled very well. Also the subtraction can remove unwanted effects, like AC Stark shift due to the cooling laser. With help of the cycling transition $c$ and the re-pumps $r_1$ and $r_2$ the transitions $s$ indicated in Fig 2.3 a) can be measured.

Transitions starting from the $^{2}S_{1}$ ($F = 3/2$) can be measured similarly by using, instead of the cycling transition, the $\Lambda$ transition shown in Fig. 2.3 b), which indicates the ion to be partly in the $F = 3/2$ ground state. One re-pumping laser is sufficient in this case. These two schemes allow to measure all the hyperfine structure splittings of the $^{2}P$ level. To have an additional knowledge about the hyperfine structure of the $^{2}S$ level, a third measurement scheme has to be applied where the transitions $^{2}S_{1}$ ($F = 1/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 1/2$) and $^{2}S_{1}$ ($F = 3/2$) $\rightarrow$ $^{2}P_{2}$ ($F = 1/2$)
2. SPECTROSCOPY ON SIMPLE ATOMIC SYSTEMS

create a $\Lambda$ system used for the shelving. This way also transition starting from the $2^3S_1 (F = 1/2)$ state can be measured. The additional re-pumping laser works on the $2^3S_1 (F = 5/2) \rightarrow 2^3P_2 (F = 3/2)$ transition.
3

Fundamentals of ion trapping

This chapter introduces the theory necessary to understand how and why an ion trap confines charged particles and how one can decrease the temperature to improve spectroscopic measurements. This provides a level of control that is not accessible in other systems.

First successful experiments with trapped ions date back to the late 50’s, when Wolfgang Paul introduced an extension of the well known quadrupole mass spectrometer to confine particles in 3D with RF fields \[2\]. Since then the field of ion trapping has been steadily growing involving more and more groups. Ion traps, from relatively simple mass spectrometers evolved to powerful tools for modern quantum optics from non-classical light sources \[29\] to quantum simulators of solid state Hamiltonians \[30\].

According to Earnshaw’s theorem it is impossible to create a potential minimum in three dimensions with only static electric or magnetic fields \[31\]. To circumvent this restriction, two main tools were developed: the above mentioned Paul traps with time-dependent electric potentials and Penning traps employing a combination of static electric and magnetic fields \[1\]. The proper design of a trap produces a field free region which, if combined with the concept of laser cooling of bound particles introduced by Wineland and Dehmelt \[32\] (following a proposal of Hänsch and Schawlow for free atoms \[33\]), leads to a very promising system for precision measurements. The first successful demonstration was published shortly afterwards, in 1978, by Neuhauser et. al. \[34\] and Wineland et. al. \[35\] who for the first time demonstrated that electromagnetic radiation can be indeed used to reduce thermal energy. During the
3. FUNDAMENTALS OF ION TRAPPING

Figure 3.1: **Paul and Penning traps** - (left) Paul trap employing radio frequency electric fields. Hyperbolical shaped electrodes produce almost ideal harmonic potential, (right) Penning trap with static electric and magnetic fields.

Over years the concept of laser cooling evolved and the new technique of resolved sideband cooling pushed the limits into the motional ground state to the $\mu$K range, allowing us to think of ions as promising building blocks of quantum computers, as proposed by Cirac and Zoller [36].

Paul’s original design relied on hyperbolically shaped electrodes that create a field free, point-like potential minimum where one ion can be stored free of perturbations associated with the trapping fields. For certain applications, like e.g. frequency standards or precision spectroscopy, it is desirable to store more ions, which are not disturbed by the environment. Prestage et. al. [37] proposed a modification of a Paul trap, the so called linear quadrupole trap, which extends the field free region to a line. It was achieved by arranging four cylindrical electrodes in a square and connecting two of them to the RF voltage while grounding or DC-biasing the remaining ones. This structure provides a time-averaged confining quadrupole potential in the radial direction with respect to the electrodes. Two rings at the ends of the rods biased with a DC field provide axial confinement (Fig. 3.2).
3.1 Linear Paul trap

The most general time-dependent potential of a 3D ion trap can be written as

$$\varphi(x, y, z, t) = \frac{1}{2}(U_{DC} + U_{RF} \cos(\Omega t)) \cdot (\alpha x^2 + \beta y^2 + \gamma z^2),$$

(3.1)

where $U_{RF}$ is the voltage amplitude of the radio frequency $\Omega$ applied to one pair of opposing electrodes and $U_{DC}$ is the DC-voltage applied to the other pair. Imposed by the Laplace equation $\Delta \varphi = 0$, restrictions on $\alpha$, $\beta$ and $\gamma$ lead to $\alpha + \beta + \gamma = 0$. For a 2D quadrupole linear trap it corresponds to $\alpha = -\beta$, $\gamma = 0$, where free motion is possible along the $z$ axis. The negative sign in the first equation means that simultaneous confining potentials in $x$- and $y$-direction is at one instant of time not possible. However it will turn out that stable trajectories are still possible, as a consequence of a time-averaged confining pseudopotential. The equations of motion decouple with respect to different radial directions so it is possible to restrict the problem to one dimension. For a particle of mass $m$ and charge $Q$, Newton’s equation gives:

$$\ddot{x} = -\frac{Q}{m} \frac{\partial \varphi}{\partial x} = -\frac{Q}{m} (U_{DC} + U_{RF} \cos(\Omega t))x$$

(3.2)

For hyperbolic electrodes $\alpha = 1/r_0^2$, where $r_0$ is the distance from the trap axis to the closest electrode surface. Introducing the following dimensionless parameters

$$a = \frac{4QU_{DC}}{mr_0^4\Omega^2}, \quad q = \frac{2QU_{RF}}{mr_0^4\Omega^2}, \quad \xi = \frac{\Omega t}{2}$$

(3.3)
it is possible to reduce the equation to the Mathieu type
\[
\frac{d^2x}{d\xi^2} + [a - 2q \cos(2\xi)]x = 0.
\] (3.4)

The \(a\) and \(q\) parameters characterize amplitudes of the static and dynamic part of the quadrupole field while \(\xi\) measures the time in units of the radio frequency cycle.

From the Floquet theorem [38] it is known, that periodic solutions of the Mathieu equation for any pair \((a, q)\) can be expressed in the form
\[
x(\xi) = A e^{i\beta \xi} \sum_{n=-\infty}^{\infty} C_{2n} e^{i2n\xi} + B e^{-i\beta \xi} \sum_{n=-\infty}^{\infty} C_{2n} e^{-i2n\xi}.
\] (3.5)

In the general case the coefficient \(\beta\), so called ”characteristic exponent”, and \(C_{2n}\) cannot be obtained analytically. Usually they are found as a recursion relation. They depend only on \(a\) and \(q\), \(A\) and \(B\) describe initial conditions. Stable solutions, hence stable ion trajectories, exist only for non-integer, real \(\beta\). Figure 3.3 shows the lowest stability region, commonly used in ion trapping experiments. For \(a = 0\) stable trajectories exist for \(q\) up to 0.9.

If we limit ourselves to \((|a|, q) \ll 1\), the characteristic exponent can be approximated by
\[
\beta \approx \sqrt{a + q^2/2}.
\] (3.6)

For many practical applications this approximation works very well, since linear ion traps are usually operated with the DC voltage \(U_{DC}\) set to zero and low enough RF amplitude so that \(a \approx 0\) and \(q \ll 1\). With this, the lowest order of the general solution of Eq. 3.5 can be simplified to
\[
x(t) \approx x_0 \cos(\omega_{sec}t)[1 - \frac{q}{2} \cos(\Omega t)],
\] (3.7)

where \(x_0\) depends on the initial conditions and
\[
\omega_{sec} = \frac{\beta}{2} \Omega
\] (3.8)

is the secular frequency. It can be interpreted as the oscillation of the particle in a confining 2D harmonic pseudo-potential formed by time-averaging over many cycles of the trap frequency \(\Omega\).

For practical purposes \(a \approx 0\), therefore
\[
\omega_{sec} = \frac{q}{2\sqrt{2}} \Omega
\] (3.9)
Figure 3.3: First stability region for a linear ion trap (red) - Yellow shows the range of parameters that give stable trajectories in $x$ directions, blue - in $y$. The overlapping region (red) means trapping both in $x$ and $y$ direction.
3. FUNDAMENTALS OF ION TRAPPING

The motion of an ion in the RF potential can be decoupled into a fast motion at a trapping frequency $\Omega$ (micromotion) which is slowly modulated at frequency $\omega_{\text{sec}}$ (secular motion). For ions created close to the trap axis the secular motion has smaller amplitude than for those created further away. Due to the high energy associated with an oscillatory motion (few eV or $10^4$ K), the initial velocity of an ionized atom does not influence the dynamics. The micromotion amplitude rises when ions move away from the axis in their secular motion (Fig. 3.4).

In the trap presented above the axial motion is not bound. Therefore to confine ions also in the axial direction, a DC voltage $U_0$ is applied to the end caps forming a potential well in which the ions arrange. The axial potential then reads

$$\Phi_{\text{axial}}(x, y, z) = U_0(z^2 - \frac{1}{2}(x^2 + y^2)) = \frac{m}{2Q}\omega_z^2(z^2 - \frac{1}{2}(x^2 + y^2)).$$

(3.10)

The ions oscillate in this potential with an axial secular frequency frequency

$$\omega_z = 2\pi\sqrt{\frac{2QU_0}{m}}.$$ 

(3.11)

![Figure 3.4: Ions trajectories in a trap](image-url)

- Trajectories of Mg$^+$ (blue, dotted) with $q = 0.15$ and Li$^+$ (green, solid) with $q = 0.51$ calculated for trap settings allowing simultaneous trapping of both species. Higher $q$’s lead to the more chaotic motion. The behavior for different distances (40 $\mu$m and 200 $\mu$m) from the axis at a time ($T = 0$) when an ion is created shows, that the closer to the axis the process takes place the smaller is the amplitude of the secular motion.
3.1 Linear Paul trap

The presence of an axial confinement influences the radial potential felt by an ion and therefore its movement can be described in terms of a "reduced" secular frequency

\[ \omega'_{sec} = \sqrt{\omega_{sec}^2 - \frac{1}{2} \omega_z^2}. \]  

(3.12)

However, neglecting the influence of an axial potential on the radial secular frequency is well justified for typical voltages applied to the trap during our experiments because the ratio \( \omega_z/\omega_{sec} \) is usually smaller than 1/10. For example for an RF amplitude of 68 V and an axial voltage of 20 V we have \( \omega_z/\omega_{sec} = 23\text{kHz/}896\text{ kHz} = 3\% \) and \( \omega_z/\omega_{sec} = 13\text{kHz/}261\text{ kHz} = 5\% \) for Li\(^+\) and Mg\(^+\), respectively.

3.1.1 Field deviation from the ideal Paul trap

An ideal harmonic potential of a Paul trap can be created only by hyperbolic electrodes. Due to the problems with machining such shapes and because of better optical access, cylindrical rods became a standard substitution. When the rods' diameter is 1.484 \( r_0 \) (\( r_0 \) is the distance from the trap axis to the surface of the electrode) they create a field that approximates the harmonic potential the best. Nevertheless, it is necessary to quantify how other arrangements of rods influence the trapping potential, especially if one thinks of more open designs like presented in the work of Maiwald et. al. [39] where an optical access of up to 96\% of 4\( \pi \) solid angle was reached.

As introduced in the previous section, at a given instant of time the trapping potential of an ideal ion trap reads \( \phi(x) = \phi_0 + \frac{U_{RF}}{2}x^2 \), if we limit ourselves to one dimension only. For a non-ideal trap, on the other hand, the potential including anharmonicities can be expanded as \( \phi'(x) = \phi'_0 + c_2x^2 + c_4x^4 + \ldots \). The polynomial \( \phi'(x) \) is even due to the reflection symmetry of an ion trap. It is now possible to define a trap efficiency \( \kappa \) as a ratio of \( c_2 \) to the coefficient standing next to \( x^2 \) in the harmonic potential: \( \kappa = 2r_0^2c_2/U_{RF} \), which gives the voltage loss factor \( L = \kappa^{-1} \). With this, the "old" stability parameters \( a \) and \( q \) transform to \( \kappa a \) and \( \kappa q \). For traps with four non-hyperbolic rods, \( \kappa \) is usually around 1 and the generated potentials are to a good approximation harmonic close to the axis.
3. FUNDAMENTALS OF ION TRAPPING

3.2 Laser cooling

Trapping is the first step towards the goals we are planning to achieve in the experiment. The high initial energy of the trapped particles (corresponding to $\sim 1000$ K) is enhanced to few eV due to the motion in the pseudo potential leading to Doppler broadening of the atomic transitions. Moreover due to the large secular orbits the ions spend most of the time in regions with large perturbing trapping fields. At the same time the interaction times with interrogating lasers are short, leading to time of flight broadening. All these effects make any precision measurements difficult. Therefore we apply laser cooling [32] to reduce the kinetic energy of the ions, damp the micromotion amplitude, localize them close to the trap axis, and hence decrease their exposure to the trapping field.

Due to the first-order Doppler effect, an ion moving towards the illuminating radiation, tuned below the resonance ("red detuned") of a cycling transition, has a higher probability to absorb a photon than an ion moving away. One absorption-emission cycle consists of the absorption of a photon of momentum $p_{\text{abs}} = \hbar k$ in laser beam direction and subsequent spontaneous re-emission. The latter process is on average isotropic ($<\Delta p_{\text{em}}> = 0$) so the net momentum transfer to the ion is $-\hbar k$ and leads to the reduction of the ion’s velocity. For a particle trapped in an ion trap one cooling beam at non-zero angle to all trap axes is sufficient. Nevertheless, it does not lead to cooling to zero momentum. The emission is a random process so an atom experiences a random walk in momentum space with $<p_{\text{em}}^2> \neq 0$. The following discussion refers only to this kind of situation, when the limiting energy is set roughly by the energy of spontaneously emitted photon, in a process called Doppler cooling.

The secular motion at the frequency $\omega_{\text{sec}}$ is the result of a pseudo-potential generated by the time-averaged trap potential 3.1. The steady state can be achieved, when the average (per cycle) cooling power $<P_{\text{cool}}>$ equals the heating power due to the spontaneous emission $<P_{\text{heat}}>$. For small velocities, i.e. in the last stage of cooling, the cooling force can be linearized and together with $T = m <v^2>/2k_b = m v_0^2/4k_b$ it gives the temperature [40]

$$T = \frac{\hbar \Gamma}{8k_b} (1 + \xi)((1 + s) \frac{\Gamma}{2\Delta} + \frac{2\Delta}{\Gamma} ).$$  \hspace{1cm} (3.13)
For the detuning $\Delta = -\sqrt{1 + s\Gamma}/2$ the minimum temperature $T_{\text{min}}$ is reached

$$T_{\text{min}} = \frac{\hbar \Gamma}{4 k_b} (1 + \xi) \sqrt{1 + s}. \quad (3.14)$$

In the above equations $m$ is the ion’s mass and $\xi = 2/5$ is a factor that takes into account the non-isotropic emission of a dipole radiation. Eq. 3.14 gives the minimal temperature of Doppler cooling to be slightly higher than $T_{\text{min}} = 0.35 \hbar \Gamma / k_b$, towards small saturation parameters $s \to 0$. A typical order of magnitude is mK. For $^7\text{Li}^+$ cooled on the $1s2s^3S_1(F = 5/2) \to 1s2p^3P_2(F = 7/2)$ transition the Doppler limit is $64 \mu\text{K}$ ($\Gamma_{\text{Li}} = 2\pi \times 3.8 \text{ MHz}$) whereas for the $^{24}\text{Mg}^+$ D2 line it equals $700 \mu\text{K}$ ($\Gamma_{\text{Mg}} = 2\pi \times 41.5 \text{ MHz}$). In a realistic situation, these limits cannot be reached mainly due to RF heating and a heating introduced by the electrodes’ material itself. Collisions with the background gas and non-zero intensity of the cooling laser also play some role but are significantly lower [41]. Nevertheless, one achieves a reduction of Doppler broadening by several orders of magnitude.

### 3.3 Mg$^+$ as a coolant for Li$^+$

Direct cooling of lithium has a lot of disadvantages, especially if it comes to spectroscopy. A cooling beam is usually much stronger than a spectroscopy beam. As a consequence, spontaneous emission and stray light will usually lead to strong background, which cannot be filtered out when, as in our case, the wavelength of the transition under study is close to or identical with the cooling wavelength. Additionally, the only accessible cooling transition in Li$^+$ starts from the metastable $2^3S_1$ ground state. We expect only of the order of 10% of the lithium ions in this triplet ground state [20] and, additionally triplet ions will decay to the singlet ground state. Only a small fraction of the ions will thus be cooled while all others will provide a hot background leading to strong heating due to the Coulomb interaction. For our purposes we envision a modified version of a spectroscopic scheme based on sympathetic cooling together with spatially-resolved detection, developed by M. Herrmann et al. [42].

The idea is based on the sympathetic cooling between ions. Loading the trap with a mixed ion chain, favorably in the configuration Mg$^+$-Li$^+$, allows cooling only a group of magnesium ions at the one end, because via Coulomb interactions the lithium ions on the other end are cooled as well. The method eliminates the temperature variations...
of the lithium ions due to the spectroscopy beam and also enables filtering out a strong cooling beam at the wavelength 280 nm with an interference filter. It also makes the crystal insensitive to whether lithium ions are in a metastable or in a ground state allowing studies of mixed metastable crystals. The presence of a cooled cloud of ions might also open the possibility of exciting already trapped ground state lithium with an electron gun without the need of reloading the trap.

There are some drawbacks of this solution like e.g. the higher expected temperature of lithium due to the almost 10 times higher Doppler limit for Mg$^+$, nevertheless it is not expected to be a limiting factor of the proposed scheme. Some technical difficulties arise from the fact, that for two species two distinct laser systems (UV and green, see 4.3) are required and optical elements have to be coated for two wavelengths. This makes the setup more complicated and more expensive. The next issue is that the stability parameters $a$ and $q$ have to be adjusted such that stable trajectories will be present for both species. The ratio of masses $\frac{24\text{Mg}^+}{7\text{Li}^+}$ equals 3.4 and from the stability diagram (Fig. 3.5) it can be seen, that the stability regions also differ by the same factor. Nevertheless, Figure 3.5 shows that both lithium and magnesium ions can be trapped simultaneously for a rather wide range of $a$ and $q$. 
Figure 3.5: Stability diagram for $^{24}\text{Mg}^+$ and $^7\text{Li}^+$ - Red - stability region for $^{24}\text{Mg}^+$, yellow - for $^7\text{Li}^+$. The overlap (dark yellow) shows the region, where both species are trapped. The stability regions for all species look similar since they depend only on $a$ and $q$, therefore to extract useful information the plot for lithium was rescaled by dividing the values for magnesium by the mass ratio 3.4. This way the correspondence between $q$ for $\text{Mg}^+$ and $\text{Li}^+$ for the same settings of the trap can be easily seen.
3. FUNDAMENTALS OF ION TRAPPING
This chapter contains a detailed description of one of the major achievements of this thesis: a working ion trap apparatus that was fully characterized using trapped magnesium ions. The apparatus is therefore ready for trapping of metastable lithium ions. The decay of metastable ions to their ground state limits the laser cooling efficiency and unless it is possible to easily re-pump them back to the metastable state they are hard to work with. For that reasons trapping and cooling of Li$^+$ to prepare it for precision measurements is not necessarily going to be as straightforward as for other species that have been already trapped. Due to the expected difficulties in handling lithium the common method of using the trapped species to test and calibrate the setup did not look promising, therefore a second species, magnesium, was trapped in the same setup. The choice was mainly governed by the availability of the laser system in the lab and the variety of literature devoted to this species.

4.1 Vacuum system

Most modern experiments provide a great level of control over the interactions between the investigated system and the environment. Usually this is achieved by setting them to work under certain vacuum conditions. For some applications, like high-harmonic generation, easily achievable pressures on the order of $10^{-6}$ mbar are sufficient. For other applications, involving, for example, ensembles of cooled atoms trapped in magneto-optical traps (MOTs), the ultra high vacuum regime is necessary to reduce interactions with the background gas and hence to prevent atoms from being kicked
4. THE APPARATUS

out of the trap.
Since the potential depth of an ion trap is orders of magnitude larger than that of a
typical MOT, for many ion trap applications the background gas is not the limiting
factor. Nevertheless ultra high vacuum helps significantly when the ultimate goal of the
experiment is to work with short-lived species or perform sophisticated spectroscopic
measurements. For lithium, high background pressure leads to the formation of LiH+.
When such "dark ions" appear during the measurement, the amount of fluorescence is
changed, which disturbs the data. Even more important is that collisions reduce the
lifetime of the metastable state. This makes long measurements impossible. The same
thing happens for magnesium, where MgH+ is created. Clear gain in terms of reduced
collisional quenching and chemical reactions can be reached by a careful design of the
system and a systematic approach to the creation of ultra high vacuum conditions.

4.1.1 Chamber

The key element of the system is a stainless steel chamber (Fig. 4.1). Access to the
interior is provided by fused silica windows which are anti-reflection coated for Mg+
and Li+ wavelengths at 280 nm and 548 nm respectively (Fig. 4.2a). For electric DC
connections to the trap, two 10-pin coaxial feedthroughs are used. The power supply
for the Channeltron and the radio frequency signal are applied via a 3-pin, high voltage,
feedthrough.

The fluorescence light is collected through a fused silica re-entry window (Fig. 4.2b).
It consists of a tube of about 25 cm length that reaches into the vacuum chamber and a
window port at the end so that the viewport is mounted as close as 35 mm to the trap
center. This allows to keep optics elements of the imaging system outside the vacuum.
The pressure readout is done with a vacuum gauge suitable for pressure measurements
between $10^{-3}$ and $10^{-12}$ mbar.

4.1.2 Pumps

Evacuating the chamber is done in four steps. First, an oil-free scroll pump (Varian
SH-110) reaches a pressure $\approx 10^{-2}$ mbar creating an environment for the turbo pump
(Leybold Turbvac 360). It decreases the pressure further down to about $5 \times 10^{-9}$
mbar. At this stage the limited ability of the turbo pump to pump out light species
requires using an ion getter pump. In order to avoid these species entering the chamber
Figure 4.1: Experimental setup - view of the setup
4. THE APPARATUS

Figure 4.2: Vacuum viewports properties - a) reflection curve for the window coatings (delivered by the manufacturer, Laseroptik GmbH) and b) re-entry window drawing

through the turbo pump the chamber is closed with a valve, the turbo pump is switched off letting the ion pump (Varian Star Cell) improve the vacuum. This way reaching pressures of about $5 \times 10^{-10}$ is possible within 1-2 days. The last step is to use a titanium sublimation pump (TSP). Flashing the TSP produces a layer of titanium on the walls and particles stick there during collisions. The procedure described above leads to the ultra high vacuum only if the chamber is baked, while the turbo pump is still running. More details can be found in the next section.

4.1.3 Preparation of parts and baking procedure

Ultra high vacuum requires specific materials. Plastic or rubber works fine down to around $10^{-6}$ mbar but the regime we are interested in constrains us to few materials that, unfortunately, are sometimes hard to machine and expensive. It is very important not to confuse zinked screws with stainless steel ones because even one, small element made of the inappropriate material can produce never ending outgassing, preventing us from reaching the desired conditions.

Conducting elements are made of copper and tantalum. The main material for the trap electrodes, mounts and screws is non-magnetic stainless steel. Except screwing and using UHV compatible connectors the only allowed connection method is welding. Glues and solder cannot be used.

As insulator, especially for parts that have to be elastic (like wires), Kapton is used. For bigger elements Macor works very well. It is relatively cheap and thanks to its
good thermal properties provides excellent base structure for atom ovens.

Once proper materials are chosen, the elements need to be cleaned. Machined parts usually are contaminated with oils, therefore they should be cleaned with special soap, e.g. Edisonite Super. A successful procedure is as follows:

- Cleaning for few tens of minutes (even hours for newly machined stainless steel parts) in an ultrasonic bath with soap dissolved in water
- Rinsing, first with running water to remove the soap and then immediately with distilled water
- Drying with a blow dryer
- Ultrasonic bath in isopropylene (methanol also can be used)
- Rinsing with distilled water
- Drying with a blow dryer

Everything above requires using powder free gloves to avoid contact with the skin, which is always a source of grease. To protect cleaned elements from dust it is a good solution to wrap them in aluminium foil which should be removed only for the assembly. When everything is connected it should be shielded with aluminium foil again to reduce contamination with dust particles which can cause some outgassing when the elements are put into vacuum. Notwithstanding clean room environment is not necessary and if handed with care a typical laboratory provides sufficient cleanness.

After closing the chamber the evacuating procedure can be started. Because of the contact of the setup with air and trace impurities inside the used materials, it outgasses a lot of water and nitrogen. To clean it, the setup must be baked. It is done by attaching heating stripes to the chamber, then wrapping it with aluminium foil. The temperature should be increased slowly, at a rate of about 1°C per minute (due to the windows’ thermal expansion). Uniformity of the temperature should be controlled mainly to avoid unwanted stresses in the chamber which can lead to cracks and therefore leaks.

At the same time the ion getter pump has to be baked. For that purpose it is sufficient to use the internal heater built into the pump. Also a current of around 35 A should be run through the titanium filaments as well as through the electron guns and ovens.
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(usually 1-2 A is sufficient) in order to bake them. A successful procedure spans a period of a few days at temperatures of around 200°C. Accordingly, cooling cannot be too rapid. However, in practice switching off all the heaters ensures a safe cooling rate in our setup.

Even though the procedure accelerates outgassing it does not stop it. The analysis of the residual gas with a residual gas analyzer Stanford Research RGA 100 shows, that even at $5 \times 10^{-11}$ mbar the partial pressure of some of the components can still be quite high. As shown in figure 4.3 peaks deriving from water ($M=2$, $H_2^+$; $M=18$, $H_2O^+$; $M=32$, $O_2^+$) and from molecular hydrogen $N_2^+$ prove, that outgassing is present despite the overall pressure reaching $10^{-11}$ mbar. Regular flashing of the titanium sublimation pump (every few days, at least) is necessary to maintain good vacuum conditions.

![Figure 4.3: Residual gas scan](image)

**Figure 4.3:** Residual gas scan - Partial pressures of the residual gases are shown, with peaks associated to most common species. The rest gas analysis was performed after baking, at the pressure around $5 \times 10^{-11}$ mbar

4.1.4 Results

An efficient procedure for achieving ultra high vacuum was established. As an effect a UHV system able to reach $5 \times 10^{-11}$ mbar was assembled.
4.2 Tank circuit

Ion traps are usually operated at radio frequencies ranging from a few up to tens of MHz. Accordingly, the applied voltages range from few tens to several hundred volts. Providing such voltages directly from a signal source and amplifier is not straightforward. Additionally, a RF ion trap has to be thought of as a capacitive load, which in general will not be impedance matched with the source and therefore reflect most of the power. Both problems are solved by using a resonant parallel LC circuit with the trap as capacitor.

4.2.1 Design guidelines

4.2.1.1 RLC circuit

In a realistic situation, ohmic losses must be included. Therefore tank circuits are modeled by a resonant RLC circuit consisting of an inductance \( L \) (coil), a capacitance \( C \) (trap) and the resistance \( R \) (wires, trap electrodes, feedthrough). The signal source is connected to a single loop of wire, movable around the coil, so that it is inductively coupled to the circuit. A circuit diagram is shown in figure 4.4.

Two main parameters characterizing the circuit are the resonant frequency \( \omega_0 = 2\pi f_0 \)

\[
\omega_0 = \sqrt{\frac{1}{LC} - \left(\frac{R}{2L}\right)^2}
\] (4.1)

and the quality factor \( Q \) describing the ability of the circuit to store energy

\[
Q = 2\pi \frac{\text{stored energy}}{\text{dissipated energy per cycle}} = \frac{\omega}{\Delta \omega} = \frac{1}{R} \sqrt{\frac{L}{C}}
\] (4.2)

where the last identity is true only when the power source is disconnected. The \( Q \) value can be easily determined, in the impedance matched case (critical coupling), where the relation \( Q = 2Q_{\text{crit}} \) holds [43]. From the above equations the voltage enhancement \( \eta \), that is the ratio of the voltage across the trap \( U_2 \) to the input voltage \( U_1 \), can be derived. In the steady state, the stored energy \( W_s \) is constant and reads

\[
W_s = \frac{1}{2} CU_2^2.
\] (4.3)

The condition for impedance matching is that the dissipated power equals the input power \( P_{\text{in}} = U_1^2 / 2Z \). It gives the dissipated energy per cycle
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\[ W_d = P_m \tau = \frac{U_1^2}{2Zf_0}. \] (4.4)

It leads to the voltage enhancement

\[ \eta = \frac{U_2}{U_1} = \sqrt{\frac{Q_{\text{crit}}\omega_0 L}{Z}} = \sqrt{\frac{\sqrt{4L^2 - CLR^2}}{4CRZ}}. \] (4.5)

The equations show that to obtain high \( \eta \), ohmic losses as well as the trap’s and circuit’s capacitance should be minimized.

**Figure 4.4: RLC circuit and coil’s dimensions definition** - (left) Simplified scheme of the resonant RLC circuit connected to the signal source of impedance \( Z \) and (right) definition of the parameters used in the coil design: \( \alpha = l/D \), \( \kappa = d/s \)

### 4.2.1.2 The coil

In most cases the quality factor of the coil will limit the total quality of the circuit, which is the reciprocal sum of the \( Q \) factors of all components \( \frac{1}{Q} = \sum_i 1/Q_i \). Therefore, to a very good approximation, all measurements of RLC circuits made of standard capacitors and resistors give, in fact, the coil’s quality. This does not have to be true when we use a trap as a capacitor.

Reducing dissipation of the energy in a coil requires optimization of the ratio \( L/C \) (Eq. 4.2). This problem was solved theoretically in [44], giving optimized values for the coil dimensions \( \alpha = l/d = 0.7 \) and \( \kappa = d/s = 0.6 \) (for definitions see Fig. 4.4). The results suggest the use of short and loosely wound coils. The next hint comes from empirical formulas for the quality factor and the inductance of coils

\[ Q_{\text{emp}} = \approx 26.5D\sqrt{f} \] (4.6)
4.2 Tank circuit

\[ L_{\text{emp}} = \approx \frac{\pi \mu_0 D n^2}{46} \]  

(4.7)

where \( \mu_0 \) is magnetic permeability of the vacuum, \( D \) the coil diameter in centimeters, \( f \) the frequency in MHz and \( n \) the number of windings. With this information we decided to use a coil with 17 windings made out of 1.5 mm silver wire and separated by 3 mm. A single loop of wire used to inductively couple the RF signal into the circuit is attached to a micrometer screw. This solution allows precise positioning of the loop and a smooth adjustment of the coupling, which is monitored with a directional coupler. When reflections are minimized impedance matching is optimized.

**Coil characterization**

The characterization of the coil is done by attaching a capacitor of known capacity to the coil to form the \( LC \) circuit. The resonance frequency is then measured using a network analyzer connected to a small loop that inductively couples to the considered circuit. With a frequency \( 2\pi \times 3.071 \) MHz and capacity 150 pF, the formula \( \omega = \frac{1}{\sqrt{LC}} \) gives an inductance \( L = 17.9 \pm 0.2 \) \( \mu \)H. If coupling is in the weak regime\(^1\) the measurement of the FWHM of the resonance line gives the quality factor of the coil \( Q_{\text{coil}} = 362 \).

For comparison: the above quantities calculated from equations 4.6 and 4.7 equal, respectively, \( Q_{\text{emp}} = 334 \) and \( L_{\text{emp}} = 17.9. \)

**4.2.2 Characterization of the tank circuit**

The coil was then connected to the trap held in the vacuum environment creating an RLC circuit. A directional coupler is used to measure the signal reflected by the trap for impedance matching between the RF signal source and the circuit. At the resonance \( \omega_0 = 2\pi \times 7.124 \) MHz and with the measured FWHM the (critical) quality factor of \( Q_{\text{crit}} = 140 \) was determined. With the given \( L = 17.9 \) \( \mu \)H, the capacitance of the trap

\[ C_{\text{trap}} = \frac{1 - 1/16Q_{\text{crit}}^2}{L\omega_0^2} \]  

(4.8)

\(^1\) in practice, weak coupling means, that the quality factor of the circuit no longer depends on the coupling. In other words, the driving force exerted on the circuit by an external source approaches zero.

\(^2\) For a second built coil with resonance frequency \( 2\pi \times 8.731 \) MHz, \( L_{\text{emp}} = 2.26 \) \( \mu \)H (measured \( 2.22 \pm 0.02 \) \( \mu \)H) and \( Q_{\text{emp}} = 321 \) (measured 292): \( \alpha = 0.78 \) and \( \kappa = 0.375 \).
and the circuit’s resistance

\[ R = \frac{L\omega_0}{\sqrt{Q_{\text{crit}}^2 - 1/16}} \]  

(4.9)

were calculated to be \( C_{\text{trap}} = 27.4 \, \mu\text{H} \) and \( R = 2.2 \, \Omega \). Having in mind that \( Q = 2Q_{\text{crit}} \), also the quality factor \( Q = 280 \) and the voltage enhancement \( \eta = 47 \) (Eq. 4.5) were extracted.

### 4.2.3 Results

**Figure 4.5: Stability parameter \( q \) versus atomic mass of a singly ionized particle**

- For the first stability region trapping is achieved for \( q \) values between 0.1 and 0.9 when \( a = 0 \). The figure shows the behavior for different RF signal source amplitudes.

Our tank circuit, with resonance frequency \( \omega_0 = 2\pi \times 7.124 \, \text{MHz} \) and \( Q = 280 \) became a very robust element of the setup. A relatively high voltage enhancement \( \eta = 47 \) removes the need of employing additional amplifiers. Moreover, with standard signal sources reaching 10 \( V_{pp} \) (in our case: Agilent 33220A), the trap is capable of trapping singly ionized particles with atomic masses between 1 and 85 (see Fig. 4.5).
4.3 Laser systems

For simultaneous trapping of Mg\(^+\) and Li\(^{++}\) two, in this case very distinct laser systems are needed. The cooling transitions for singly charged magnesium and lithium ions are in the UV and green, respectively, therefore it is necessary to use fused silica optical elements, preferably with anti-reflective coatings for both wavelengths.

The hyperfine structure of Li\(^+\) complicates the measurement of the fine structure. On the other hand it can provide additional knowledge of the nuclear structure of lithium. Especially extracting the isotope shift between \(^6\)Li\(^+\) and \(^7\)Li\(^+\) is of great interest for precision nuclear size measurements which leads to a better understanding of the halo nuclei \(^{11}\)Li\(^+\) [25].

The anticipated measurement of the complete fine- and hyperfine multiplet with the shelving scheme described in Chapter 2 requires one or two fixed-frequency lasers on the cycling or Λ transitions and for repumping. To access the transitions in a frequency range of about 170 GHz, a tunable and narrow-band spectroscopy laser is also necessary. As these requirements are hard to fulfill with one laser together with acusto- and electro-optic devices, we plan to use two different lasers for the Li\(^+\) spectroscopy.

4.3.1 Magnesium II laser system

The UV light is shared with another experiment on \(^{24}\)Mg\(^+\). It is produced in bow-tie second harmonic generation cavities (SHG) and has to be sent then in free space to the lithium setup over about 5 meters.

Photoionization laser

For photoionization of neutral Mg laser light at 285 nm is required. 570 nm light is created with a CW dye laser (CR699/21) pumped with a Coherent Innova-400 argon ion laser or a Coherent Verdi V10. The light is sent via a single mode fiber to a resonant SHG cavity. The cavity is stabilized to the laser frequency using the well-known Hänsch-Couillaud polarization lock scheme [45]. Typically 200 mW of input power are converted in a BBO crystal resulting in about 1 mW output power at 285 nm. The beams exiting the BBO have distorted shapes, with interference patterns present. Shaping is done with two cylindrical lenses behind the cavity. Spatial filtering of the beam improved its quality and significantly reduced divergence. The photoionization beam has to travel
in a free space more than 5 m, therefore a lens compensating divergence was added. The frequency of the photoionization laser is monitored with a commercial wavemeter (Angstrom WS Ultimate 2).

Cooling laser

For cooling, the D2 transition \((3s_{1/2} - 3p_{3/2})\) at 280 nm is employed. This frequency can conveniently be generated from an all-solid-state CW fiber laser amplifier system and two frequency-doubling stages [46]. A fiber laser (Koheras) at 1118 nm delivers 2 W output power with a linewidth well below 1 MHz. The light is then frequency-doubled twice in resonant SHG cavities similar as for the photoionization laser. In the 1st stage 400 mW at 560 nm are generated in an LBO crystal. The second stage uses a BBO to generate 10 mW near 280 nm.

Although this turn-key laser system is very convenient, the fiber laser was not operational during our measurement. We replaced it by a CR 699/21 CW dye laser (Rhodamine 19) at 560 nm and frequency-doubled the light in the BBO stage of the original fiber laser setup. 400 mW at 560 nm resulted in typically 10 mW at 280 nm. After shaping with cylindrical lenses, the cooling light is overlapped with the photoionization light using a polarization beam splitter cube and the two beams are then focused into the trap.

4.3.2 Lithium II laser system

The cooling transition between the metastable state \(1s2p^3S_1(F=5/2)\) and \(1s2p^3P_2(F=7/2)\) at 548.5 nm is easily accessible with commercial lasers.

Dye laser

The laser light for the cycling and \(\Lambda\) transition as well as for repumping needs to be rather powerful. As described in Chapter 2, various different beams have to be created using electro-optic and acousto-optic modulators. For this purpose we use a second Coherent 699/21 dye laser pumped by a Coherent Innova 400 argon ion laser in the visible multiline mode (mainly emitting at 514 nm and 488 nm). A 532 nm laser cannot be used in this case as this pump wavelength is too close to the desired output wavelength of 548 nm. The dye laser we use, with Rhodamine 110 dissolved in ethylene
glycol, allows tunability around 550 nm over the range of more than 10 nm [47]. The dye laser output power reaches up to 20% of the pump power. For the needs of the experiment several hundred mW is sufficient.

The laser is equipped with a temperature-stabilized low finesse Fabry-Perot cavity for short-term stabilization. The control bandwidth of the commercial laser is limited to a few kHz without additional measures. As a result, a typical linewidth of about 1 MHz is achieved. This is well bellow the fine und hyperfine structure splittings so that the transitions can be addressed separately. However, it is of the order of the natural linewidth of the Li\textsuperscript{+} 2\textsuperscript{3}S-2\textsuperscript{3}P transition and would thus limit ultimate precision in the spectroscopy.

**Diode laser**

For spectroscopy we need a laser that covers a broad frequency range (more than 170 GHz over the whole HFS multiplet). Moreover it should have large control bandwidth in order to achieve a narrow laser linewidth. As the saturation intensity of the Li\textsuperscript{+} 3\textsuperscript{S}-3\textsuperscript{P} transition is only $I_s = 63$ W/m\textsuperscript{3}, the laser powers required are rather low. When focussed to 0.5 mm, 12 µW is sufficient.

The requirements are well fulfilled by a diode laser. We use a commercial frequency-doubled Toptica TA-SHG10 laser with an output power of 7 mW at 548 nm. It has a coarse tuning range of more than ±1 nm and can be mode-hop-free tuned over 20 GHz. Its linewidth is specified to 1 MHz (in 1 ms) but can be significantly reduced due to its high modulation bandwidth of up to 1 GHz.

### 4.4 Ion storage and detection

This section contains a detailed description of the elements that are directly responsible for creation, trapping and detection of ions. Initially, trapping of ions was performed with a trap that was lent us by Hans Schüssler from Texas A&M University. However, it turned out that the large insulator pieces in this trap were significantly charging up when using the electron gun over several hours. The increasing potentials quickly deteriorated and eventually hampered trapping at all. Thus we decided to change to a trap design that minimizes insulator surfaces close to the trapping volume.
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4.4.1 Ion creation

4.4.1.1 Atomic ovens

![Figure 4.6: Lithium and magnesium ovens.](image) - (left) lithium and (right) magnesium oven. The metal shields with holes, aligned with the dispensers, serve as collimators reducing the divergence of the atomic beams and preventing the trap electrodes from being coated.

Lithium oven

As a source of lithium atoms commercially available dispensers from Alvatec are used [48]. The dispenser’s tube is 3 mm in diameter and contains about 20 mg of natural lithium (92.58% $^7$Li, 7.42% $^6$Li). The tubes are filled with argon and sealed with indium in order to prevent oxidation of lithium. Due to the technical difficulties in handling lithium it is impossible to get sources smaller than 2 mm in diameter. This leads to the high temperature inertia of the dispenser.

The base of the oven is made out of Macor (Fig. 4.6, left). A V-groove along the Macor piece ensures proper alignment of the dispenser with a hole in a shield in front of the oven.
4.4 Ion storage and detection

Figure 4.7: Lithium source activation and temperature-current calibration. (left) to access lithium, the indium sealing has to be melted. First, around 2 A current melts indium and leads to argon release (peak). To evaporate lithium, the current is increased to about 8 A (after [48]). (right) temperature-current calibration of the dispenser done under $10^{-2}$ mbar pressure.

**Magnesium oven**

Thin natural magnesium (abundance $^{24}$Mg$^+$ 78.99%, $^{25}$Mg$^+$ 10.00%, $^{26}$Mg$^+$ 11.01%, [49]) wires are available, therefore the oven is significantly smaller than for lithium (Fig. 4.6). A 0.5 mm wire is put inside a tantalum tube, which is heated.

The wire requires some special treatment before it is used in vacuum. The magnesium oxide layer, which forms in air, has a higher boiling point than magnesium itself hence it is possible, that liquid magnesium can erupt when the temperature is increased. To prevent this the wire has to be put in citric acid to remove the magnesium oxide. Instead, a magnesium citrate layer is then created on the surface, which does not interact with air and melts at lower temperatures than magnesium.

**4.4.1.2 Electron gun for creation of metastable $^7$Li$^+$**

The $^2P$ states of two-electron systems are of major interest for theoreticians. To probe them it is necessary to start from the metastable $^2S$ level. To do that one has to excite ground state singly ionized lithium by providing 59 eV of energy, which corresponds to 21 nm. The only efficient sources that work at this wavelength can either be found in big research facilities like DESY or are bulky and very expensive. Moreover this extreme ultraviolet wavelength is not transmitted in any window material.

The most straightforward approach is to use an electron gun. However, the radio
frequency trapping fields interact with the electrons disturbing their trajectories and therefore this solution is not very accurate. Additionally it is not very efficient because the cross section for the Li→Li⁺ process is only about $10^{-22\pm1}$ cm² (well below the photoinization cross section of magnesium, $(8.1 \pm 2.3)10^{-17}$ cm², [50]). Nevertheless, a fraction of about 10% of the ions in the metastable state can be expected [20]. Two

![Electron gun](image)

**Figure 4.8: Electron gun** - Cut-away view of the used electron gun. The inset shows how the Wehnelt cylinder and filament are connected to the power supplies [43]

electron guns, of design shown in Fig. 4.8 are used: one above the trap and one on the side 4.9. Electrons are emitted from the tip of the filament (made out of 0.25 mm tungsten wire) by heating it with 5.5 A current. The exact value of the current depends on the specific filament design and can vary even between seemingly equal filaments (e.g. when the tips are bent at slightly different angles). A potential difference of several hundred volts is applied between the filament and the cathode (grounded plate with a hole in front of the electron gun) to accelerate the emitted electrons. The main drawback of using an electron gun is that it creates a lot of unwanted ion species. Indeed, it is very easy to fill the trap with background ions, even at the pressure of the order of $10^{-11}$ mbar. Additionally, the ejected electrons build up charges on insulating surfaces creating potentials, which can distort the trap.

### 4.4.1.3 Photoionization of $^{24}$Mg and $^{26}$Mg

Whenever it is possible, photoionization is done with lasers. Magnesium exhibits a two-step transition to the continuum at 285 nm. The 285 nm light excites a transition
from the ground state to the $3s3p^1P_1$ state. Subsequently, a further 285 nm photon or a 280 nm photon from the cooling beam excites the electron into the continuum. The process is partially isotope selective, because the 3 GHz isotope shift between $^{24}\text{Mg}$ and $^{26}\text{Mg}$ exceeds the photoionization laser linewidth. By tuning the wavelength, the isotope composition of the ion cloud can thus be controlled.

### 4.4.2 Ion trap

The ion trap is the most important part of the setup. It is directly responsible for trapping of ions and its design and performance is crucial for the project. Because the electron gun is an inevitable part of the experiment, the trap design was

![Figure 4.9: Ion trap](image)

- Two views of the trap with accompanying equipment: Channeltron (on the top, above the central region), two electron guns (on the top and on the side) as well as Mg and Li ovens (under the trap and on the side, respectively). Ions are created in the outer region to protect the central parts of the electrodes against contamination, which can distort the trapping potential. (left) view from the side of the lithium oven, (right) view from the imaging system side

mainly governed by the need to reduce the surface of the insulators so that they do not charge up and to hide them from the trap volume. In addition with the need of having segments the end caps are constrained to a ring-shaped design, which implies the drawback of using high voltages (even above 1 kV) if tight axial confinement is necessary. Kapton, instead of Macor, was used as a material for insulators, mainly because it is flexible and does not break under stress, which makes it relatively easy to machine. Rods and mounts are made from stainless steel. The trap was originally
designed by Sebastian Knünz as a part of his PhD and later adopted for the lithium setup.

The trap is a 4-rod linear Paul trap. Two rods are connected to the radio frequency signal while the remaining two are grounded (Fig. 4.10). Four outer rods support the trap’s structure and additionally can be used for micromotion compensation. All electrical connections inside the vacuum chamber are done with Kapton wires (copper with an insulating layer of Kapton). The RF wire is placed as far as possible from other elements to minimize capacitance. The dimensions are shown in Fig. 4.10. The design provides much better optical access than both the original Paul trap and an ideal quadrupole trap allowing higher detection efficiency.

The trap is not an ideal hyperbolic Paul trap, therefore the potential it creates is not strictly harmonic. To account for the deviations, the voltage loss factor \( L = \kappa^{-1} \) was calculated (see Sec. 3.1.1). The radial potential was simulated with software package Comsol for a given instant of time, both for x-direction (between RF rods) and y-direction (between grounded rods). The function \( \phi_i = \phi_{0,i} + c_2 x_i^2 \), where \( x_i \in \{x, y\} \) was fitted to the obtained data (see Fig. 4.11). Due to numerical limitations, the fields in both directions slightly differ but still give essentially the same \( \kappa \) and \( L \) equal 0.82±0.01 and 1.22±0.02, respectively. It implies, that the applied voltages have to be 1.22 times higher than for an ideal trap to achieve the same quadrupole potential in the center of the trap. Figure 4.12 shows a potential along the z-axis. Peaks correspond to the position of the rings. Fitting is done only for the central region, which is destined for trapping.
The power of an ion trap as a tool for precision spectroscopy comes from the possibility of confining ions in a field-free region. However, in practice unwanted stray fields can occur, shifting an average ion position from the node of the AC potential leading to micromotion and, as a result, RF heating. There are three main sources of these fields [51]:

- **phase difference between the trap electrode potentials** - caused by asym-
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...metries in the electrical impedances of the electrodes. For example, a phase shift will occur if the leads to the trap electrodes have different inductances due to different lengths or geometrical arrangements.

- **coating of the electrodes** - an effusive oven may unevenly coat the trap electrodes with its content, which could cause contact potentials of a fraction of a volt. Additionally, the trap electrodes may become unevenly charged when this coating or other dielectric or oxide layers are charged by the emitted electrons.

- **patch effects** - different crystal planes at the surface of the electrodes can also produce surface potential variations of roughly 100 mV.

Although the magnitude of the latter two can be reduced by heating the trap electrodes *in situ*, no techniques can eliminate these fields. The ion’s transition may therefore be distorted by a second-order Doppler shift (time dilation) or AC Stark shift. To minimize micromotion, a DC voltage has to be applied to two of the outside rods. By varying its magnitude and sign it is possible to shift ions to the node of the RF potential.

### 4.4.3 Channeltron for destructive detection

An efficient way of detecting charged particles is using active layers, which release electrons when hit by a particle (Fig. 4.13, left). Common devices used in ultra high vacuum environments are Channel Electron Multipliers. In the experiment, a Channeltron 4839 model from Burle Industries Inc. is used. By applying a high voltage...
(typically -2.8 kV) to the front part of the device, positively charged ions are accelerated onto the active layer and generate electrons. These are accelerated by the same voltage towards the rear, grounded part. When hitting the active layer, they release secondary electrons. Eventually an electron avalanche is generated that causes a measurable macroscopic current pulse for each impinging ion. To limit the influence of the high accelerating voltage on the trap, the entire device, except the front part, is shielded with a metal sheet housing. The front part has a steel mesh built in, which is set to several hundred volts.

It was observed, that also atoms emitted from the oven create a detectable signal. This is usually easily overcome by taking measurements only after the oven’s shutdown. Unfortunately, contamination of the active layer decreases the performance of the Channeltron. Also UV light can create photoelectrons, therefore while working with magnesium the UV beams should be blocked during the measurement since the presence of the UV beam can increase the background counts by 3 orders of magnitude.

4.4.3.1 The q-scan method

A q-scan is a low resolution mass spectrometry method done with a Channeltron. It is especially useful, when ions not addressed with lasers need to be detected. It is also an excellent tool to check if the trap is operating and get a rough estimation on the species trapped and their numbers.

No quantitative theory exists for this type of detection. Qualitatively however it is well understood. The stability of ions is characterized by the stability parameter \( q = \frac{2QU_{RF}}{mr^2\Omega^2} \) which is proportional to the trapping voltage amplitude and is inversely proportional to the ion’s mass. When the voltage is ramped down, the q parameter changes accordingly and at some point the ion trajectories are no longer stable. They are then released from the trap, with heavier ions escaping first because they become unstable before lighter species do. This results in peak(s) seen in the Channeltron’s signal.

The method is certainly not suitable for precision measurements but is an invaluable tool when setting up a new ion trapping experiment.
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4.4.3.2 Notch filter

The presence of radio frequency electric fields strongly distorts the Channeltron signal by picking up the trap driving frequency. For sufficiently high trapping potentials as used in the experiment, the amplitude of a picked-up RF signal can reach several hundreds of mV leading to the signal disappearing under these oscillations. Therefore no reliable detection would be possible. To overcome this problem a passive notch filter was built (Fig. 4.14 left). Despite the simple design it proved to be a very robust solution providing a RF signal reduction of about 24 dB (see Fig. 4.14 right).

![Notch Filter Diagram](image)

*Figure 4.14: Notch filter @7.324MHz* - (left) the circuit diagram of a notch filter. A trimmer is used to precisely tune the resonance frequency of the notch filter to the trap driving frequency. (right) Comparison of the signal from the Channeltron with (red line, solid) and without (black, dotted) the notch filter. All other features of the setup are unchanged. The trap frequency was $2\pi \times 7.324$ MHz, the trigger level -10 mV, as for all measurements with the Channeltron.

4.4.4 Imaging system for non-destructive optical detection

For laser-cooled ions like Mg$^+$ a convenient way of non-destructive detection can be achieved by observing the photons scattered during laser cooling. When shining laser light on the cycling or $\Lambda$ transition of Li$^{++}$, also these ions can be detected optically. In our setup photons are to be collected simultaneously by two detectors (Fig. 4.15): an Andor iXon+885 EMCCD camera and a photomultiplier Burle C31034A built in a PHOTOCOOL PC182CE housing. Both are cooled to $-80^\circ$C and $-30^\circ$C respectively.
to reduce dark counts. The image of the ions by an objective is split by a beam splitter and directed to both detectors. In order to avoid expensive coatings for two wavelengths a special holder is used, which allows changing between two sets of beam splitters together with appropriate filters, one for Mg$^+$ at 280 nm and one for Li$^+$ at 548 nm. The camera gives spatially resolved information about the ions and allows taking pictures of the cloud. Spatially resolved detection also allows to extract the signal from single ions. The drawback of the CCD camera is its slow readout, which is limited to about 30 frames/sec at full resolution. The photomultiplier has a higher quantum efficiency ($\sim 70\%$ instead of $\sim 25\%$@280 nm and $\sim 90\%$ instead of $\sim 55\%$@548 nm [53, 54]) and provides fast readout, therefore is more suitable for spectroscopy.

Typical distances between trapped ions are on the order of 10 $\mu$m. The 1004$\times$1002 pixels chip of the CCD camera we are using has a pixel size of $8 \times 8 \mu$m. Therefore to capture pictures with resolved ions the image has to be magnified so that the image of the ion-ion distance spans several pixels. The imaging is accomplished by one objective formed by two 2-inch bestform lenses, yielding a total focal length of 76.5 mm. It gives a magnification of -11 for a distance from the trap center to the closest lens of 83.2 mm. The magnification is mainly limited by the distance of the objective to the trap center, which cannot be smaller than 70 mm due to the design of the re-entry window. The solid angle covered by a cone of opening $\theta$ reads $\Omega = 4\pi \sin \theta / 4^2$. The
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83.2 mm distance from the trap center to the closest lens makes the objective cover about 33.9°, corresponding to 2.2% solid angle. When illuminated at a saturation parameter $s=1$, the maximum count rate per Mg$^+$ ion is around 378 kHz and around 61 kHz per Li$^+$ ion. The above estimations include 65% transmission of a Schott UG-11 UV Edge filter, 90% transmission of a beam splitter and 70% detection efficiency of a photomultiplier at 280 nm and 90% transmission of an interference filter, 90% transmission of a beam splitter and 90% detection efficiency of a photomultiplier at 548 nm.

For some applications like studies of mixed crystals it could be desirable to know precisely the relation between pixels and real distances in a trap. Calibration of the length scale can be done by loading the trap with two magnesium ions and measuring the ion-ion distance $\Delta x'$ in pixels and the center-of-mass (secular axial) frequency $\omega_z$. From that, the conversion factor from pixels to meters as well as the magnification of the imaging system and resolution can be calculated. Following [55] the separation of two ions of a charge $Q$ and mass $m$ confined in a potential with secular frequency $\omega_z$ can be derived to be

$$\Delta x = \sqrt[3]{\frac{e^2}{2\pi\epsilon_0 m \omega_z^2}}.$$  \hfill (4.10)

The image size on the camera’s chip equals $x[\mu m] = p_x \times \Delta x'$ where $p_x$ is the pixel size in $\mu m$. To achieve better accuracy for the calibrated values, the above mentioned equations are used to derive a function, that is fitted to the data

$$\Delta x' = \sqrt[3]{\frac{e^2}{2\pi\epsilon_0 m p_x^2} M \omega_z^{-2/3}},$$  \hfill (4.11)

where $M$ stands for magnification. Different values of $\omega_z$ and thus $\Delta x'$ are achieved by changing the axial voltage since $\omega_z$ relates to this voltage through equation 3.11. Analysis of a signal from the camera allows to determine the distance between the centers of the ions. Exemplary data are shown on Fig. 4.16 (courtesy of S. Knünz). Because the light distribution is approximately gaussian, this curve is fitted to a cross section of an ion spot on the camera. From the FWHM the resolution can be calculated.
Figure 4.16: Examplary calibration data for an imaging system - For illustration of the method the function 4.11 is fitted to the data points taken for different axial voltages by Sebastian Knünz in another experiment in the lab. The resulting magnification is 93, resolution 2.6 μm and correspondence 1px=0.532 μm.
4. THE APPARATUS
5

The results

The following Chapter presents results, that are crucial for the further course towards laser spectroscopy of lithium ions. It was not only proved that the setup is capable of trapping ions, but the trapping of magnesium allowed us to characterize the setup in all details and compare the results with previously calculated values. In particular, secular scans provide valuable information for the characterization of the imaging system and the tank circuit.

5.1 Enhancement of the tank circuit

In secular scans an additional RF potential is applied to the end caps or to one of the micromotion compensation rods. The ions’ secular motion can be excited axially and radially, respectively, and hence the ions are heated. For crystallized ions it leads to the crystal’s melting when the external fields reach one of the secular frequencies of the motion. This can subsequently result in ions escaping the trap.

The consideration leading to the design of the tank circuit allowed to determine a theoretical value of the circuit’s voltage enhancement. Secular scans provide a method that can be used to confirm the calculation with the actual value. From the section 4.2 it is known, that the trap’s voltage $U_{RF}$ can be written as $U_{RF} = \eta U_{SG}$, where $U_{SG}$ is the voltage amplitude generated by a signal generator and $\eta$ is the voltage enhancement. With the equations 3.3 and 3.9 the voltage enhancement reads

$$\eta = \sqrt{\frac{2\omega_{sec}m\Omega_{RF}r_0^2}{2QU_{SG}}}.$$  (5.1)
5. THE RESULTS

To exploit the above equation it can be re-written in a form that is convenient for fitting

\[ \nu_{sec} = 4484\eta U_{SG}, \]  

(5.2)

where the numerical factor was calculated for magnesium ions and for a trap frequency of \( 2\pi \times 7.329 \) MHz and \( \nu_{sec} = \omega_{sec}/2\pi \).

The measurement procedure is as follows: the trap is loaded with an ion cloud, which is then crystallized by laser cooling. An external RF voltage is applied to one of the micromotion compensation rods to assure radial excitation of the crystal. While scanning over the secular frequency, the fluorescence from the ions is being observed. At resonance the ion crystal melts and a signal drop is observed. All steps are repeated for different trap voltages \( U_{SG} \). At the early stage of the experiment the secular frequency \( \omega_{sec} = 2\pi \times 643 \) kHz for 1.6 V RF amplitude applied to the tank circuit was measured. The received value \( \eta = 45 \) was in a reasonably good agreement with the value \( \eta = 47 \), calculated based on the tank circuit analysis with the network analyzer. Due to an equipment failure more detailed measurements, following the above procedure, were not performed.

5.2 Destructive detection with the Channeltron

![Figure 5.1: Q-scan of the background gas](image)

The peak is a confirmation that the trap can confine particles. The relatively large width is a result of the high temperature of the ions. (right) double peaks due to multiple species confined by the trap. The horizontal axis is in the units of percentage of the initial voltage, which is ramped down from 100% to 0%.
Before the imaging system was set up and aligned, it was necessary to confirm if the trap worked. The only way of doing that was to use the Channeltron. The software we used presents data in the way, that the first point is 100% of the initial voltage and the last one is 0%. First, the electron gun was run for about a minute creating ions out of the background gas. Trapping was enhanced by cooling of background gas ions by helium, used as a buffer gas at a pressure of $5 \times 10^{-7}$ mbar. During that period the trap was on and set to the RF voltage supporting stable trajectories for species heavier than 20 a.u. (atomic units). After switching off the electron gun we waited 20 seconds and the helium line was closed. When after about 10 seconds the helium pressure decreased to $10^{-9}$ mbar we performed a q-scan measurement which showed a peak (see Fig. 5.1). A good consistency check of the trap operation is to repeat the q-scan for a different trap voltage. An increase of the initial voltage resulted in a peak moved to the left (towards lower percentage). This is in agreement with the expected behavior as it means that ions were ejected for the same RF voltage. It is well known, that even a relatively good vacuum has significant amounts of N\textsubscript{2} outgassing from the walls, therefore the peaks can most likely be assigned to this species. This is supported by the mass spectrometry measurement of the background gas, done with a residual gas spectrum analyzer, which showed a significant partial pressure of the N\textsubscript{2} (see Fig. 4.3). It was the first prove, that the trap is working the way we expected. Similar scans were done with magnesium (see Fig. 5.2). With the oven switched on but without any ionization sources the q-scan was flat. Next, when the oven was on and the photoionization beam was sent into the trap creating ions, a q-scan resulted in a peak. Repeating the procedure with cooling and photoionization beams showed a slight increase in the number of detected particles due to the more efficient ionization (two step process, described earlier in the thesis). When only the cooling beam was present atoms evaporating from the oven could not be ionized and the signal was again flat. Because no other species could have been ionized with a 285 nm light, magnesium was proven to be indeed trapped.

5.3 Optical detection of magnesium ions

Once the operation of the trap was established, optical detection of the Mg\textsuperscript{+} ions was anticipated. As the detected light comes from the cooling transition of a $^{24}$Mg\textsuperscript{+} ion,
Figure 5.2: Q-scans for magnesium - a) oven on, no ionization sources, b) oven on, photoionization on - ions are created and trapped, c) oven on, photoionization on, cooling beam on - ions are created and cooled, slight increase mainly due to the enhancement of the ionization by the cooling beam, d) oven on, photoionization off, cooling beam on - no signal, magnesium cannot be ionized, therefore the 280 nm light does not produce fluorescence.
5.3 Optical detection of magnesium ions

Figure 5.3: First images of ion cloud and ion crystal - Top-one of the first captured images of an ion cloud, bottom-one of the first ion crystals created in our trap. The distorted shape is due to the imaging system.

the alignment of the cooling beam is critical for detection. If it does not hit the cloud directly on the axis, the fluorescence is very weak or vanishes completely. Fluorescence detection turned out to be extremely sensitive to the alignment, even though apertures were placed to provide repeatable beam positioning.

For large negative detuning of the cooling laser from the center of the transition (≈ 50 MHz) the ions were seen as a cloud of plasma (Fig. 5.3, top) with typical temperatures of several hundred milikelvin. When the resonance was approached from the red side the ions experienced a phase transition, which resulted in a great increase of the emitted light. The ions form cigar-shaped, crystal-like structures arranged in shells, with single ions visible at the tip of the crystal (Fig. 5.3, bottom). The shape of an ion crystal (as well as a cloud) is mainly a result of a balance between Coulomb repulsion and compression due to the trapping potential. Additional effects are present, like RF heating of outer ions. The high micromotion magnitude leads to the transfer of energy from the rf drive into the random motion of the ions through their mutual Coulomb interaction. The kinetic energy associated with the micromotion can be several orders of magnitude higher than the thermal energy at which spatial ordering occurs in static
5. THE RESULTS

Figure 5.4: Ion crystals - $^{26}$Mg$^+$ crystals for increasing RF amplitude (from the top of the figure). Empty spaces in the string (bottom) are due to $^{24}$Mg$^+$

potentials, so the coupling of micromotion into thermal motion can be expected to be critical for crystal formation. An increase of the RF amplitude leads to the crystal’s squeezing in the radial direction. With weak axial confinement it is possible to reduce a crystal to a string. Fig. 5.4 shows the result of this process. Ions in the outer shells are exposed to RF heating, so ions located there create more blurry images in comparison with the ones located on axis, where the micromotion amplitude approaches zero. With the above pictures it was confirmed that the setup works the way we expected. Even more important is that the resolution and the overall quality of the imaging system were achieved with inexpensive bestform lenses.
Summary & Outlook

The construction of the apparatus is the first step of the long term goal aiming at measuring the fine structure and other interesting properties (see Chapter 2.) of metastable lithium ions. In our first trap, major problems arose from large Macor pieces charging up when the electron gun was running. Although trapping of electron-impact ionized species from residual gas was achieved, the trapping conditions were not sufficiently reproducible and stable for spectroscopy. In our new trap this problem has been solved by a design using smaller insulator pieces and hiding them behind conducting elements, so that possible remaining charges would not disturb the trapping fields. A first characterization of the trap was performed with Mg\(^+\) generated by photoionization. Laser cooling has been implemented to create ion crystals that will serve as coolants for embedded Li\(^+\) ions. Even though it was not proved that our setup can trap Li\(^+\), successful trapping of photoionized magnesium as well as trapping electron-impact ionized species from the residual gas give us a strong evidence, that it would be also possible for lithium ions. It remains to be investigated what fraction of the lithium ions can be generated in the metastable state.

In summary, in the framework of this thesis the following goals were achieved:

- an ultra-high vacuum system reaching pressure down to \(5 \times 10^{-11}\) mbar was assembled

- an ion trap that is largely insensitive to charging up due to the electron guns and a tank circuit with high voltage enhancement of \(\eta = 47\) were built and successfully taken into operation.
6. SUMMARY & OUTLOOK

- diagnostics of the ion trap was performed with different methods:
  - q-scan
  - secular scans
  - electron gun loading of the trap from the background gas
  - photoionization loading of Mg$^+$
  - crystallization of Mg$^+$
  - optical detection of Mg$^+$ with single ion resolution

- further directions leading to the efficient detection and creation of lithium ions were explored. The application of the charge exchange process for metastable lithium creation was proposed.

Outlook

The experience we gathered while building the experiment, gave us a deeper insight into what kind of problems we may expect once lithium ions are trapped and forced us to think ahead of alternative approaches that had been not expected, when the project started. This chapter presents some of the necessary steps that have to be made towards trapping of lithium as well as additional ideas, that can be investigated in the framework of future experiments.

6.1 Creation of Mg$^+$ crystals with electron impact ionization

In our setup magnesium ions play a double role: they are used as a coolant for Li$^+$ and serve as a guinea-pig for preliminary tests of all the components of the setup. Even though it can be easily photoionized a procedure of efficient loading of the trap with one of the Mg$^+$ isotopes using an electron gun has to be established. Mg$^+$ is detected optically so that the influence of the electron gun on the ions can be monitored in real time, which is not necessarily the case for Li$^+$. The influence of the RF fields on the electron beam makes a precise adjustment of electrons' energy impossible, therefore the optimal value has to be determined experimentally. A first approach in our new
6.2 Trap content purification

trap, with accelerating voltages up to 500 V, was unsuccessful. However, electrons did not seem to influence the trap content, leaving the trapped Mg\(^+\) crystals unperturbed. In most experiments involving this species, electron energies were around 1 keV (see e.g [56]), which shows, that the correlation between ionization energy (7.65 eV) and the optimum electron energy is not very obvious. This makes the studies of electron impact ionization of magnesium a very valuable benchmark for the operation of the electron guns before switching to lithium.

6.2 Trap content purification

Secular scans are considered as a very powerful tool for purifying the trap content. It is not of utmost importance when atoms are photoinized, since this process is usually at least partially isotope selective and does not influence the background gas. When electrons are used for ionization (and excitation) it is necessary to remove unwanted species, which are confined in a trap together with the ions of interest. In a considered scenario magnesium is photoinized and cooled while metastable lithium ions are created with an electron gun. This leads not only to the filling of the trap with Mg\(^+\), Li\(^{+*}\) and Li\(^+\) but also with Li\(^{2+}\), Li\(^{3+}\) and the residual gas ions. The first two are essential for the experiments, the presence of the others leads to heating which may be strong enough to make sympathetic cooling due to the Mg\(^+\) ions inefficient. Tuning the RF voltage applied to one of the micromotion compensation rods to the frequency of a secular motion of the unwanted species induces resonances leading to heating and expelling the ions from the trap. This way it is possible to get rid of the rest gas ions, leaving the trap only with species required for the experiment. This method does not remove Li\(^+\), because it is indistinguishable from Li\(^{+*}\), as far as its secular frequency is considered. Nevertheless, it promises a great improvement in terms of a purity of the trap content. The mass resolution of the method presented above is sufficient to achieve efficient purification. When applied to magnesium ions it is possible to remove \(^{24}\text{Mg}^+\) and \(^{25}\text{Mg}^+\) leaving the trap only with \(^{26}\text{Mg}^+\).

\(^1\text{Sebastian Knünz, private communication}\)
6.3 Alternative detection of lithium ions

Metastable lithium ions can be detected optically but the expected low number of ions in this state might cause difficulties at the early stages of the experiment, before all parameters are optimized. Therefore it is crucial to have an independent way of efficient detection of Li$^+$ without relying on the optical transition. It would not only allow controlling the progress of the experiment but, what seems to be even more important, would give an additional confirmation that the trap is loaded with Li$^+$. When this is confirmed it is easier to focus on the search for the fluorescence from Li$^{+*}$. One of the possible detection schemes would be a q-scan. The drawback of this solution is, that in its simpler version it mainly confirms, that "something" was trapped but does not give a definite answer of what was the exact content of the trap. The proposed approach combines a Channeltron-based detection with secular scans. First, the trap is loaded with $^{24}$Mg$^+$ which is then crystallized. Subsequently, an RF voltage is applied to one of the micromotion compensation rods and its frequency is scanned. Fluorescence is measured with a photomultiplier and simultaneously a signal from the Channeltron is recorded. When the applied RF crosses the resonance with the secular frequency of $^{24}$Mg$^+$, the crystal melts into a plasma, which is indicated as a fluorescence drop. When heating is absent, the plasma re-crystallizes and fluorescence returns to its previous level. For large enough amplitudes at least some ions should escape the trap resulting in a peak in the Channeltron signal. The same procedure is repeated for $^{26}$Mg$^+$. With this it is possible to determine the secular frequencies for both isotopes and associate them with peaks in a Channeltron signal. Therefore with two points it is possible to calibrate the RF frequency scale for the Channeltron signal using the equation

$$A = \frac{\kappa QU_{RF}}{\sqrt{2m\Omega br^2_0\omega_{sec}}}.$$ 

where $A$ denotes the atomic number. For typical trap settings supporting stable trajectories for Mg$^+$ and Li$^+$ the secular frequencies of both magnesium isotopes are around 200 kHz, separated by $\approx 4$ kHz. From [57] it is known, that the resonances for magnesium have a width of a few kHz so based on the above equation the expected width of the peak in the Channeltron signal ("mass width") should not exceed 0.5 atomic units. For lighter species secular frequencies are higher, therefore even if the resonance is wider than for heavy ones it should give a narrow "mass width".
advantage of this method comes from the fact that species with atomic masses 5, 8, 9 are not expected to be created with an electron gun so they should not be present in the spectrum and therefore even wide "mass width" would allow to confirm whether Li\(^+\) has been trapped or not.

### 6.4 Laser-assisted charge transfer for Li\(^{+*}\) creation

Beginning with early work on lithium ions (see e.g. [19]), Li\(^{+*}\) had been usually created by electron impact ionization of a lithium atomic beam or by some variation of this method, like discharge ionization of LiCl [26] or other lithium compounds [58]. For trapping Li\(^+\) in an ion trap, electron impact seems to be a very promising tool, which proved to be relatively efficient in experiments with other species and molecular ions. Nevertheless, this approach is not very efficient in creating metastable ions. An expected fraction of metastable Li\(^+\) of around 10\% may be reached [20] but it does not assure us, that for our applications this solution is the best available. It certainly has a lot of drawbacks and requires trap content purification to remove unwanted ions created out of the residual gas. Therefore an additional approach could be to utilize laser assisted charge exchange, which is common for plasma physics [59]. This idea is briefly discussed below.

Two colliding particles under certain conditions can exchange electron(s) in a process $A + B^+ \rightarrow A^+ + B^*$. The corresponding charge transfer cross section is known

![Figure 6.1: Charge exchange in an atomic collision](image)
6. SUMMARY & OUTLOOK

to decrease rapidly with the energy defect $\Delta E$ between the donor state of atom $A$ and the acceptor state of atom $B$. The cross section for non-resonant charge transfer (Fig. 6.1 b) is generally very small but approaches $10^{-14}$ cm$^2$ in the accidental case of resonance $\Delta E = 0$ (Fig. 6.1 a). It was shown, that in the presence of intense light the cross section for the non-resonant process can approach that of the resonant process if the energy $h\omega$ of the incident light approximately equals the energy defect $\Delta E$ between donor and acceptor states in the non-resonant process (Fig. 6.1 c,d). For our goal the base ion would be Li$^{2+}$. The energy difference between its ground state and the metastable state of Li$^+$ is 16.62 eV and it can be trapped and sympathetically cooled by Mg$^+$. Simultaneous trapping of Li$^{2+}$, Li$^+$/Li$^{+\ast}$ and $^{24}$Mg$^+$ is possible, with stability parameters $q$ equal e.g. 0.4, 0.2 and 0.06 respectively. The next step would be to add neon to the vacuum chamber. With its ionization energy of 21.56 eV neon could be a donor of an electron in a resonant charge transfer if the energy defect $\Delta E = 4.94$ is compensated by a laser light. This corresponds to photons of a wavelength of 251 nm. Radiation sources at these wavelengths are commercially available, therefore no technical difficulties are expected. The process Li$^{2+}$+Ne$+h\nu \rightarrow$Li$^{+\ast}$+Ne$^+$ is expected to convert Li$^{2+}$ into Li$^{+\ast}$ with an efficiency reaching 100%. The last step would be to remove the trapped Ne$^+$. This can be done e.g. by continuous application of an additional RF signal resonant with the secular frequency of Ne$^+$ such that once neon ions are created they are immediately heated and expelled from the trap. Currently it is not clear whether this approach will be necessary or at least advantageous in a real setup over the direct creation of Li$^{+\ast}$ by electron impact, nevertheless this direction could be worth investigating in the future.
References


REFERENCES


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