Implementation of a Coherent Lyman-α Source for Laser Cooling and Spectroscopy of Antihydrogen

by

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Abstract

This dissertation describes two related projects: the development of a coherent Lyman-\(\alpha\) source and the implementation of a supersonic hydrogen beam.

A two-photon resonance-enhanced four wave mixing process in krypton is used to generate high power coherent radiation at \(\omega_{Ly-\alpha} \Rightarrow 121.56\) nm, the hydrogen Lyman-\(\alpha\) line, to perform spectroscopy and cooling of magnetically trapped antihydrogen (\(1s - 2p\) transition). This is a tool to directly test both the Einstein Equivalence Principle and Charge, Parity, and Time inversion symmetry. The former can be tested by measuring the gravity interaction of matter and antimatter. Inversion symmetry can be tested by comparing the spectroscopic properties of hydrogen and antihydrogen. Both experiments require optically cooled antihydrogen. Under the current trapping conditions, optical cooling could be performed with nanosecond long pulses of 0.1 \(\mu\)J of Lyman-\(\alpha\) radiation at a repetition rate of 10 Hz.

The process to generate Lyman-\(\alpha\) radiation uses two wavelengths (\(\omega_R \Rightarrow 202.31\) nm and \(\omega_T \Rightarrow 602.56\) nm), which are mixed in a sum-difference scheme (\(\omega_{Ly-\alpha} = 2\omega_R - \omega_T\)) with a two-photon resonance at (\(4s^24p^5p[1/2]_o \leftarrow 4s^24p^6(1S_0)\)). The source implemented produces 1.2 \(\mu\)W at the Lyman-\(\alpha\) line and this was confirmed by performing spectroscopy of hydrogen. The design, implementation and characterization of the source are discussed in this dissertation.

In the second part of the dissertation the implementation of the hydrogen beam and its characterization are discussed. The atomic hydrogen is generated with a thermal effusive source and it is entrained by an expanding noble gas. This process generates a cold beam of hydrogen atoms.
Abstract

Hydrogen is separated from the noble gas with a Zeeman bender that uses the forces generated by the Zeeman shift of low field seeking states of hydrogen and engineered magnetic field gradients. The hydrogen beam was characterized with a quadrupole mass spectrometer. The seed noble gas beam was characterized by colliding it with ultra-cold rubidium atoms in a magneto-optical trap. The trapped atoms loss rate resulting from these collisions can be used to measure the density of the atomic beam. This measurement demonstrates the potential of using magneto-optical traps as absolute flux monitors.
Preface

All the work presented henceforth was conducted in Dr. Takamasa Momose’s laboratory with the exception of the work presented in Appendix A, which was performed at CERN on the ALPHA experimental apparatus.

The material presented in Chapters 2 and 3 is being prepared for publication and Dr. Momose and I are the only authors. This work has also been presented in two conferences: Canadian Association of Physicist Congress 2013 (CAP 2013) and the 11th International Conference on Low Energy Antiproton Physics (LEAP 2013). Portions of Chapter 3 have been submitted as an invited article for the Physics in Canada Magazine (due to the presentation winning a prize during the CAP 2013 Congress). Proceedings of LEAP 2013 have been accepted for publication in the journal Hyperfine Interactions (Springer). In both publications I am the first author. I was responsible for all the design, implementation and assembly of the experimental equipment and I performed all the calculations, data collection, and data analysis presented under direct supervision of Dr. Momose.

All the hardware design and implementation of Chapter 4, besides the last section (Collision scattering experiments) was done by myself with the help of two undergraduate students: Andrew Wong (undergraduate summer student) and Polly Yu (undergraduate thesis student). Some of the data collection and analysis was performed by Polly Yu under my direction and is included in this dissertation (specifically the time of flight data used for Fig. 4.11). The remaining data collection and analysis was done by myself. The implementation of the pulsed valve discussed in this chapter was done by Pavle Djuricanin, Research Technician, and Sajjad Haidar, Electronics Technician, from Technical Services in the Chemistry Department as noted. The aspects of the valve development presented here represents
all of my contribution.

The last section of Chapter 4 (Collision scattering experiments) is a collaboration effort with Dr. Kirk Madison. The magneto-optical trap (optics and light source) was mainly implemented by members of Dr. Madison’s laboratory with the help of Dr. Matthias Strebel from Freiburg University. My contribution is limited to the hardware implementation (the vacuum system and mechanical systems) and the integration with the supersonic beam. All the data collection and the analysis presented was done by myself. This work is being prepared for publication and I am the first author.

Finally, all the work presented in Appendix B, regarding the carbon nanotubes was performed in collaboration with an undergraduate summer student, Bill Wong, who I supervised. A publication is in progress from this work.
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<tr>
<td>AD</td>
<td>Antiproton Decelerator</td>
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<tr>
<td>ALPHA</td>
<td>Antimatter Laser Physics Apparatus</td>
</tr>
<tr>
<td>ASE</td>
<td>Amplified Spontaneous Emission</td>
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<tr>
<td>ATHENA</td>
<td>Antihydrogen Production and Precision Experiment</td>
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<tr>
<td>BBO</td>
<td>$\beta$-Barium Borate</td>
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<td>CERN</td>
<td>Conseil Europen pour la Recherche Nuclaire</td>
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<tr>
<td>CPT</td>
<td>Charge, Parity and Time</td>
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<tr>
<td>CR</td>
<td>Capacitive-resistive</td>
</tr>
<tr>
<td>CVD</td>
<td>Chemical Vapour Deposition</td>
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<tr>
<td>LIF</td>
<td>Laser Induced Fluorescence</td>
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<td>MOT</td>
<td>Magneto-optical Trap</td>
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<tr>
<td>QED</td>
<td>Quantum Electrodynamics</td>
</tr>
<tr>
<td>SM</td>
<td>Standard Model</td>
</tr>
<tr>
<td>SME</td>
<td>Standard Model Extensions</td>
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<tr>
<td>EEP</td>
<td>Einstein Equivalence Principle</td>
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<tr>
<td>EUV</td>
<td>Extreme Ultraviolet</td>
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<tr>
<td>FWHM</td>
<td>Full Width at Half Maximum</td>
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<tr>
<td>FWM</td>
<td>Four Wave Mixing</td>
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<tr>
<td>IGBT</td>
<td>Insulated-gate Bipolar Transistor</td>
</tr>
<tr>
<td>KDP</td>
<td>Potassium Dihydrogen Phosphate</td>
</tr>
<tr>
<td>LFS</td>
<td>Low Field Seeking</td>
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<td>MA</td>
<td>Mirror Coil A</td>
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<td>MWNT</td>
<td>Multi-walled Nanotubes</td>
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<tr>
<td>Nd:YAG</td>
<td>Neodymium-doped Yttrium Aluminum Garnet</td>
</tr>
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<td>PMT</td>
<td>Photomultiplier Tube</td>
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<td>QMS</td>
<td>Quadrupole Mass Spectrometer</td>
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<td>Resistive-capacitive-inductive</td>
</tr>
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<td>RE-FWM</td>
<td>Resonance-enhanced FWM</td>
</tr>
<tr>
<td>RIKEN-RAL</td>
<td>RIKEN at Rutherford Appleton Laboratory</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscope</td>
</tr>
<tr>
<td>SHG</td>
<td>Second Harmonic Generation</td>
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<tr>
<td>SWNT</td>
<td>Single Walled Nanotubes</td>
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<td>S1</td>
<td>Solenoid 1</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission Electron Microscope</td>
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<td>$\text{TEM}_{nm}$</td>
<td>Transverse Electro-magnetic Mode ($nm$)</td>
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<td>THG</td>
<td>Third Harmonic Generation</td>
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<td>TPA</td>
<td>Two-photon Absorption</td>
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<td>VUV</td>
<td>Vacuum Ultraviolet</td>
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<td>XUV</td>
<td>Extreme Ultraviolet (150 nm - 30 nm)</td>
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<td>$\chi^{(n)}$</td>
<td>$n$ Order susceptibility</td>
</tr>
<tr>
<td>$\vec{P}_{NL}$</td>
<td>Nonlinear polarization</td>
</tr>
<tr>
<td>$r^{\alpha}$</td>
<td>Transition dipole matrix element</td>
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<tr>
<td>$G^{(n)}$</td>
<td>Nonlinear Green's function</td>
</tr>
<tr>
<td>$\Gamma$</td>
<td>Electric transition damping constant</td>
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<tr>
<td>$\Gamma_{Bkg}$</td>
<td>MOT atom loss rate constant</td>
</tr>
<tr>
<td>$\Gamma_{R}$</td>
<td>Homogeneous Lorentzian linewidth of resonant state</td>
</tr>
<tr>
<td>$\omega_{Ly-\alpha}$</td>
<td>Hydrogen Lyman-$\alpha$ line angular frequency</td>
</tr>
<tr>
<td>$\omega_{R}$</td>
<td>Two-photon resonant laser angular frequency</td>
</tr>
<tr>
<td>$\omega_{T}$</td>
<td>Tuneable laser angular frequency</td>
</tr>
<tr>
<td>$F$</td>
<td>Phase matching constant or atom flux</td>
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<tr>
<td>$\Delta k$</td>
<td>Phase difference</td>
</tr>
<tr>
<td>$f_{ab}$</td>
<td>Oscillator strength</td>
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<tr>
<td>$S_{ab}$</td>
<td>Line strength</td>
</tr>
<tr>
<td>$S(\omega)$</td>
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<td>$A_{ab}$</td>
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<tr>
<td>$b$</td>
<td>Confocal length</td>
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<tr>
<td>$C$</td>
<td>Vector mismatch per atom</td>
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<tr>
<td>$N_a$</td>
<td>Density number of species $a$</td>
</tr>
<tr>
<td>$L$</td>
<td>Nonlinear medium interaction length</td>
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<tr>
<td>$f(\nu)$</td>
<td>Maxwell-Boltzmann distribution</td>
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<tr>
<td>$\Delta \omega$</td>
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<td>$\eta$</td>
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Acknowledgements

I would like to thank Prof. Takamasa Momose for providing me with the opportunity to get involved in this project. I would also like to thank Prof. Andrea Damascelli for his mentorship and advice. Thank you to Dr. Kirk Madison who has made the collision experiments possible. Finally, I would like to thank Prof. Jeff Young and Dr. Roman Krems for sharing their expertise.
Chapter 1

Introduction

Hydrogen Lyman-α radiation (the $1s - 2p$ transition line of the hydrogen atom at 121.56698 nm or $2.466068 \times 10^{15}$ Hz) is important because it allows for the excitation and detection of ground-state hydrogen atoms. The transition is the strongest resonance in hydrogen and has been mostly used in physical cosmology to map the density and location of hydrogen clouds (absorbers) and the location of quasars (sources) in the universe. It is also used to calculate redshifts and to extract information about universe expansion and the cosmological constant. Additionally, direct photometry of the spectral line has been used to study the Sun and Mars atmospheres[1]. Excitation with Lyman-α radiation has also been used to measure hydrogen density [2] and as diagnostic tool for fusion plasmas. A very important application of coherent Lyman-α radiation is optical cooling of hydrogen. Laser cooling, or more precisely, Doppler cooling of magnetically trapped hydrogen was first reported by Walraven et al in 1993 [3, 4]. No other hydrogen optical cooling experiment has been reported since then.

The trapping of antihydrogen (the antimatter counterpart of atomic hydrogen), recently reported by the ALPHA (Antihydrogen Laser PHysics Apparatus) collaboration at CERN (Conseil Europen pour la Recherche Nuclaire or European Council for Nuclear Research) facility, has revived interest in Lyman-α sources [5]. The ALPHA collaboration is an international group of researchers (which the author has recently become part of) whose main purpose is to achieve stable trapping of antihydrogen to study fundamental symmetries. A number of measurements have been performed or planned for the trapped antihydrogen at ALPHA such as microwave (hyperfine splitting), one-photon ($1s - 2p$) and two-photon ($1s - 2s$) spectroscopy, and gravity interferometry. The ultimate goal of these exper-
Chapter 1. Introduction

iments is to test Charge, Parity, and Time (CPT) inversion symmetry (or matter-antimatter symmetry), and antimatter-gravity interactions.

One of the difficulties in performing these measurements of the magnetically trapped antihydrogen, to the precision required for a fundamental symmetry or gravity interaction test, is its relatively high kinetic energy. Energetic antihydrogen is difficult to hold in a stable position in a trap and this leads to line broadening from the Doppler effect and from Zeeman shifts. The high kinetic energy of the antihydrogen also makes interferometry measurements of antimatter-matter gravity interactions difficult due to the poor statistical significance achievable. In addition, the antihydrogen production rate is still very low which results in very low-density samples making traditional evaporative cooling impossible. For these reasons optical cooling of the trapped antihydrogen, using the $1s - 2p$ one-photon (Lyman-$\alpha$) transition, is essential. The purpose of the current work is to develop a coherent Lyman-$\alpha$ source capable of eventually performing optical cooling of the trapped antihydrogen in the ALPHA apparatus, and the initial efforts towards this goal are described in this dissertation.

In this introductory chapter I first describe the motivation for the current undertaking (i.e. the development of a source capable of optical cooling trapped antihydrogen) and provide a short description of the experiments envisioned. Some background on antimatter and the theory behind antihydrogen and fundamental symmetries research is given to substantiate the motivation. This is the only discussion on antimatter in this dissertation. Secondly, hydrogen and its Lyman-$\alpha$ transition are discussed with emphasis on the details important for the experiment. Thirdly, the overall structure of this dissertation is described.

The remainder of the dissertation will move away from antihydrogen research and will focus on the design, implementation and characterization of the Lyman-$\alpha$ source. The Lyman-$\alpha$ radiation is generated through a nonlinear optical process and the theory needed to understand how the source works is described at the beginning of Chapter 2. The rest of Chapter 2 and Chapter 3 will go through the design, implementation and characterization of the source. Chapter 4 will describe the implementation and
characterization of a supersonically expanded beam of hydrogen atoms. An important part of the beam characterization is done through elastic collision scattering experiments of the beam atoms and ultra-cold rubidium atoms on a magneto-optical trap (MOT) (used as an absolute pressure sensor device). These experiments are described at the end of Chapter 4 and with this I conclude the dissertation.

1.1 Motivation for cooling antihydrogen

Atomic hydrogen has been one of the primary research vehicles for atomic physics that has led to the foundations of quantum mechanics and quantum electrodynamics. It has been the continuous development of the hydrogen model and metrology techniques that has made it the most precisely studied system in physics. In fact, the $1s - 2s$ spectroscopy of hydrogen is the only measurement with a relative precision below $5 \times 10^{-15}$ without laser cooling [6]. Attempts to explain its spectra have triggered the development of new theories continuously changing the physics paradigms. The Balmer formula, Balmer’s attempt to explain the hydrogen emission lines now bearing his name, provided the foundation of Rydberg’s work. The Rydberg’s formula (validated by the discovery of the hydrogen Lyman series of emission lines) established the concepts of wave number and principal quantum number. Bohr’s model of the hydrogen atom postulated the foundations of quantum mechanics. Sommerfield and later Dirac included relativistic effects to the existing hydrogen models perfecting them and introducing important physics (such as the Sommerfield’s fine structure constant and the antimatter solution of the Dirac equation). Lamb and Retherford measurements on hydrogen spectra triggered the development of quantum electrodynamics (QED) (the quantum theory of electromagnetic fields) and part of the standard model (SM) of particle physics (the currently accepted theory of subatomic particle interactions) leading to an explanation of what is known as the Lamb shift of energy levels. More recently measurements of the $1s - 2s$ transition in hydrogen have been used to test physics beyond the standard model and also what is known as the
1.1 Motivation for cooling antihydrogen

standard model extensions (SME) introduced by Kostelecký [7]. The for-
ermer test set limits on the possible temporal variation of the fine structure
constant (one of the parameters that determines the hydrogen energy lev-
eels) [6, 8] and the latter tests (SME) are searching for Lorentz and CPT
violations using two measurements separated by half of earth solar orbit
period [9]. The trapping of antihydrogen starts a new chapter on hydrogen
research. With stable trapped cool antihydrogen several new experimen-
tal possibilities become realizable. Two experiments that come to mind
are a comparison of spectroscopic properties of hydrogen and antihydro-
gen (CPT Invariance) and the measurement of the gravitational interaction
between matter and antimatter.

1.1.1 A brief introduction to antimatter and antihydrogen

Antihydrogen is the bound state of an antiproton and a positron (antielec-
tron). It is considered the simplest anti-atomic system and the antimatter
analog of hydrogen. However, this definition is only suitable to specialists
as it does not directly tell us anything about the system that we want to
study. What exactly do we mean with the term antimatter? The problem
defining antimatter comes from the fact that the term matter is perhaps
poorly defined in science. A starting point is defining matter as anything
composed of particles that have rest mass and occupy classical volume.
However, quarks and leptons are considered point particles with no effec-
tive volume. Nevertheless, volume can be argued to be the result of the
Pauli exclusion principle. In addition, a common use of the term mat-
ter refers to the energy-momentum tensor that is the source of the grav-
itational field in general relativity. This is a common definition in cos-
mology for instance. The problem with this definition is electromagnetic
fields (photons) contribute to the total energy-momentum of the system
(i.e. mass) and therefore add matter to it (isolated photons are generally
not considered matter as they have zero rest mass). For the sake of discus-
sion let us define ordinary matter as anything composed of quarks and lep-
tons. Antimatter then is anything composed of antiquarks and antileptons
1.1. Motivation for cooling antihydrogen

which are particles with the exact rest mass and spin of their corresponding quarks and leptons but with opposite sign charge. This is known as the Feynman-Stueckelberg interpretation of antiparticles. These particles have been observed and their properties have been measured to various degrees of accuracy. Table 1.1 shows a compendium of particle-antiparticle comparisons. A summary and discussion about these comparisons can be found in Ref. [10]. Ref. [11] provides an extensive list of comparisons showing no significant difference.

Table 1.1: List of particle-antiparticle property comparisons and the relative precision of the measurement. The quantities measured for each particle pair are identical within the uncertainty of the measurement. The relative precision is given by the difference between the measurements of the particle-antiparticle pair normalized by one of the measurements itself ((x_{particle} − x_{antiparticle})/x_{particle}). The uncertainty shown represents the precision of a CPT test of the specific property. If the uncertainty is not shown it was not specifically calculated in the reference.

<table>
<thead>
<tr>
<th>Particle Pair</th>
<th>Property</th>
<th>Relative Precision</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>e⁻e⁺</td>
<td>mass (m)</td>
<td>8×10⁻⁹</td>
<td>[12]</td>
</tr>
<tr>
<td></td>
<td>charge (q/m)</td>
<td>–</td>
<td>[13]</td>
</tr>
<tr>
<td></td>
<td>g-factor (g₂)</td>
<td>2×10⁻⁹</td>
<td>[14]</td>
</tr>
<tr>
<td>p̅p̅</td>
<td>mass (m)</td>
<td>2×10⁻⁹</td>
<td>[15]</td>
</tr>
<tr>
<td></td>
<td>charge (q/m)</td>
<td>9×10⁻¹¹</td>
<td>[15]</td>
</tr>
<tr>
<td></td>
<td>magnetic moment (µ)</td>
<td>3×10⁻³</td>
<td>[16]</td>
</tr>
<tr>
<td>K⁺K⁻</td>
<td>mass (m)</td>
<td>1×10⁻¹⁸</td>
<td>[17]</td>
</tr>
<tr>
<td></td>
<td>decay width (τ)</td>
<td>–</td>
<td>[17]</td>
</tr>
<tr>
<td>µ⁺µ⁻</td>
<td>g-factor (g₂)</td>
<td>7×10⁻⁸</td>
<td>[11]</td>
</tr>
</tbody>
</table>

In addition to experimental confirmation, antimatter is a consequence of quantum mechanics and special relativity. As a matter of fact, before antimatter particles were detected they were theoretically predicted. The Dirac equation predicted the positron [18, 19] a couple of years before Anderson reported its discovery on a cloud chamber in 1932 [20]. The an-
1.1. Motivation for cooling antihydrogen

tipron (composed of three antiquarks) was observed much later in 1955 at the Bevatron accelerator in Berkeley [21]. The positron solution of the Dirac equation (the relativistic Schrödinger equation) is a consequence of special relativity as it contains a very important symmetry that allows the existence of antimatter. This symmetry, strong reflection, involves the reflection of the four coordinates (spatial and time) through the origin. This reflection applied on Maxwell’s equations changes the sign of the electric charge [22, 23]. For instance, reversing the spatial coordinates of an electron moving along an electric field line reverses the electric field and the magnetic field. If time is subsequently reversed (the particle retraces its path), an observer unaware of the symmetry operations would infer that the particle is of positive charge. In Ref. [22] Feynman shows that a positron emerges from an electron when proper time is reversed. The quantum mechanical equivalent to strong reflection is called the CPT theorem. In quantum theory the equations are complex and CPT inversion takes this into account by combining strong reflection with complex conjugation. Charge conjugation C is an operation that conjugates the complex wave function changing the sign of the charge in the particle. Parity P is an operation that inverts the space coordinates. Time reversal T is an operation that interchanges the forward with the backward light cone (time inversion). According to this theory antihydrogen is the CPT symmetric reflection of hydrogen, and as such, all the physics related to hydrogen should be identical to that of antihydrogen. More precisely, the CPT theorem predicts that the positron and antiproton that compose the antihydrogen atom should have identical mass and total lifetime, but equal and opposite charge and magnetic moment as the electron and proton that compose the hydrogen atom. In fact, some of the initial measurements performed on antihydrogen such as a resonant quantum transition between hyperfine states confirm this theory [24].

Antihydrogen was first synthesized at CERN [25] in 1995 and Fermilab [26] in 1998 through collisions of relativistic antiprotons with a nucleus. In such interactions, some of the antiproton’s energy could be converted to an electron-positron pair and some of these positrons bound to an antiproton
1.1. Motivation for cooling antihydrogen

creating relativistic antihydrogen (i.e. moving at relativistic speeds). The first non-relativistic antihydrogen atoms (with energies of the order of 1 eV) were synthesized by the ATHENA experiment in 2002 at the antiproton decelerator (AD) at CERN [27]. These atoms were still too energetic to trap and escaped the apparatus in a few microseconds. The ALPHA collaboration, formed as a successor to ATHENA, observed the first signs of trapped antihydrogen in 2009 [28] followed by a definitive identification in 2010 [5]. Currently ALPHA is able to trap antihydrogen for more than 15 minutes [29]. This achievement presents a unique opportunity to further investigate the properties of antimatter, and to precisely compare matter and antimatter. The trapping of stable antihydrogen opens the door to laser cooling experiments, and with it, the possibility of high precision measurements becomes realizable. This is why the source implemented in this thesis could play a vital role.

1.1.2 Comparison of the properties of hydrogen and antihydrogen

None of the physical properties compared on matter/antimatter systems have revealed significant statistical differences (see Table 1.1) and CPT inversion symmetry has been validated in every experiment. However, antihydrogen is the fist antimatter atom (anti-atom) ever created and this opens the door for a range of new possible CPT inversion symmetry tests. For instance, any small difference between the properties of electrons and positrons or protons and antiprotons would potentially result in differences between electronic (or positronic) transitions of hydrogen and antihydrogen. This is one of the motivations for measuring the electronic transitions of antihydrogen such as the \( (1s - 2s) \) or Hyperfine transition.

To measure new physics effects at short distances, colliders usually require collisions with energy \( (E_c) \) of the same order of magnitude \( (\Lambda_{sd}) \) as the scale of the physical phenomena studied: \( E_c \approx \Lambda_{sd} \). Precision measurements at low energies access short distance scales by measuring energy...
1.1. Motivation for cooling antihydrogen

shifts that typically have the form [30]:

\[ \Delta E \sim \frac{m^{n+1}}{\Lambda_{sd}^n}, \tag{1.1} \]

where \( n \) depends on the effective field theory and is typically \( n > 0 \), and \( m \) is the characteristic energy of the system. CPT violations, as will be explained below, are expected to emerge from Planck scale (very short distance) physics (\( \Lambda_{sd} \sim 10^{19} \text{ GeV} \)). As an example, if \( n = 1 \) and \( m \sim \text{GeV} \) then the energy shifts are in the order of \( 10^{-19} \text{ GeV} \) and that would be the required resolution to observe a CPT violation. Following this argument, for example, ALPHA calculated the frequency precision required for \((1s - 2s)\) spectroscopy of antihydrogen to be \( 10^3 \text{ Hz} \) and for Hyperfine splitting to be \( 10^{-4} \text{ Hz} \) [10]. However, it can be argued that using the framework of a field theory, as above, to calculate an energy shift caused by CPT violation is unfounded since there is no formal accepted field theory that includes CPT violation. In other words, using a field theory to quantify a figure of merit for testing a CPT violation is inconsistent with the field theory since the validity of the field theory relies on the validity of the CPT theorem. The problem on testing CPT relies on it being one of the fundamental pillars of the current physics paradigm. The CPT theorem of Luders and Pauli [31] states that any local (point-particle), Lorentz invariant, and causal field theory will be symmetric under the combined transformation C, P and T. The theorem is one of the foundations of quantum field theory and the standard model of particle physics. Nevertheless, a CPT violating theory (i.e. Kostelecký’s SME) could account for a quantum theory of gravity (the string theory which the SME is based on) and for the observed Baryon asymmetry (the observed matter antimatter asymmetry in the universe) [32].

The currently accepted quantum field theory and standard model of particle physics incorporate the electromagnetic, strong and weak interactions. Gravity, on the other hand, cannot be incorporated. This has lead to the development of grand unified theories or theories of quantum gravity. In general, these theories break Lorentz invariance (and CPT symmetry). In fact, a theory of gravity (specifically string theory) served as a motiva-
1.1. Motivation for cooling antihydrogen

tion to devise a CPT and Lorentz violating extension of the SM compatible with most of the SM characteristics [33–35]. String theory rejects the idea of point-particles and replaces them with strings with dimensions of approximately one Planck length \((1/m_{\text{Planck}})\). As a result particle interactions are non-local. By extension space-time takes a non-local character and, in the theory, interactions in distances less than the Planck length do not exist. Quantum mechanical predictions on smaller scales are ignored. As explained by Kostelecký, the non-locality of string theory leads to CPT violation and Lorentz symmetry breaking [34]. However, all the elementary particle processes currently accessible occur over distances much greater than the strings. The observation of these processes is equivalent to macroscopic observations of the underlying atomic structure of matter. Macroscopic phenomena can be approximated by physics that does not necessarily take into account the quantum structure of matter, but is the result of quantum many-body interactions. Similarly, Planck-scale physics may lead to some effective continuum quantum field theory that is approximately local [36] and leads to the standard model of particle physics. Following this trend, Kostelecký described various types of interactions that would reveal CPT and Lorentz violations within the standard model extension [7, 37]. Some of the interactions can be studied in the hydrogen/antihydrogen system and a short review can be found in the review by Holzscheiter et al. [38]. In summary, it appears that ideal candidates for Lorentz and CPT violation tests are (see Fig. 1.1 for a graphical explanation of the tests):

- Comparison of the \(1s - 2s\) two-photon transition line of mixed spin states in antihydrogen and hydrogen confined in a magnetic trap with an axial bias. This transition line has a very narrow intrinsic line width (1.3 Hz) and has been precisely measured in hydrogen (the \(1s - 2s\) transition has been measured as \(2\ 466\ 061\ 413\ 187.103(46)\) kHz \([1.9\times10^{-14}\ \text{relative uncertainty}]\), using Doppler-free two-photon spectroscopy [8].

- Comparison of the ground state hyperfine transitions involving spin-
1.1. Motivation for cooling antihydrogen

flip in antihydrogen and hydrogen confined in a magnetic trap with an axial bias field. The ground-state zero field hyperfine splitting has been measured as $1420\,405\,751.7662(30)\,\text{Hz}$ $[2.1\times10^{-12}\text{ relative uncertainty}]$ $[39]$. Both of these tests could potentially be performed with the antihydrogen trapped by ALPHA. The antihydrogen in the trap has been shown to have a lifetime longer than 15 minutes $[29]$ and is within an axial bias field. Also a new trap (ALPHA2) scheduled to start operating in 2014 has been designed with laser and microwave access ports. However, The temperature of the trapped antihydrogen is still too high to perform the high-precision measurements required to see the Lorentz and CPT violation effects. The trapped antihydrogen translational energy is less than 0.5 K ($\times k_B$) and is free to roam in a volume of approximately $4\,\text{cm}^2 \times 30\,\text{cm}$ $[29]$. The non-uniformity of the magnetic field and high kinetic energy of the antihydrogen leads to large Zeeman broadening. This is currently the main limitation on microwave and laser spectroscopy $[24,41]$. A CPT and Lorentz violation test using the hyperfine transition, for instance, would require a precision of $1/3\,\text{mHz}$ $[38]$ which is currently beyond the capability of ALPHA (currently in the order of MHz) $[24]$.

This is where laser cooling of antihydrogen could play a vital role. Laser cooling would reduce the kinetic energy of the antihydrogen localizing the sample in the centre of the trap where a more uniform field is achieved. This will dramatically reduce Zeeman effects and also the second order Doppler broadening for two photon spectroscopy. In addition, having a localized cold sample opens the door to shelving strategies where one atom is recycled and the transition is repeated many times. This technique has been showed to increase the precision of the measurement.

Optical cooling (Doppler cooling) $[4]$ of magnetically trapped hydrogen was demonstrated in 1993 by Walraven et al. $[4]$ using a pulsed Lyman-$\alpha$ source. Starting with a spin-up polarized hydrogen gas with a density $n \geq 10^{11}\,\text{cm}^{-3}$, at 80 mK (pre-cooled via evaporative cooling) they were able

\[1\text{This is in contrast to laser induced evaporation which was also used by Walraven to cool hydrogen below recoil temperatures but is not what is being referred to in this work}\]
1.1. Motivation for cooling antihydrogen

Hydrogen-Antihydrogen

Cooling Transitions

\( M_F = 0 \)

\( M_F = 0 \)

\( 2S_{1/2} \)

\( 2P_{3/2} \)

\( 2P_E_{\sigma} \)

\( 82\ 258.2850014 \text{ cm}^{-1} \)

\( 0.204 \text{ cm}^{-1} \)

\( 82\ 258.9543992821 \text{ cm}^{-1} \)

\( 82\ 258.9191133 \text{ cm}^{-1} \)

\( n = 2 \)

Lyman-\( \alpha \)

\( 2\ 466\ 068 \text{ GHz} \)

Two-photon Transition

\( \pi_1 \sigma_1 \)

\( F=1 \)

\( F=0 \)

\( F=0 \)

\( F=1 \)

\( F=2 \)

\( F=0 \)

\( F=0 \)

Dirac+ Lamb Shift

+ Fine Structure

Hyperfine Splitting

Hyperfine Transitions

CPT1

CPT2

\( M_F = 0 \)

\( M_F = 0 \)

\( F=0 \)

\( F=1 \)

\( F=2 \)

\( n = 1 \)

\( n = 2 \)

Figure 1.1: Energy diagram of hydrogen and antihydrogen and the Lyman-\( \alpha \) transition for cooling. Cooling can be accomplished with the \( ^2S_{1/2} \rightarrow ^2P_{3/2} \) transitions (\( \sigma_1 \) and \( \pi_1 \)). The two possible CPT violation tests: CPT1 (hyperfine splitting involving spin flip) and CPT2 (1s – 2s two-photon transition of mixed spin states) are shown. CPT1 involves spin flip between the trapable states \( F=1 \) (\( M=0 \rightarrow M=+1 \)). It is possible to measure it using the transitions (\( \sigma_1 \) and \( \pi_1 \)) to non-trappable states. The two-photon transition test, CPT2, is between \( M = +1 \) states and, therefore, more difficult to measure. The energy levels are based on Ref. [40].
1.1. Motivation for cooling antihydrogen

to cool the hydrogen to 8 mK in 15 min while increasing the density by a factor of 16. The Lyman-α source delivered $3 \times 10^7$ photons per pulse at 50 Hz and 10 nsec pulses. This is equivalent to a power of $4.9 \times 10^{-11}$ J at 50 Hz = $2.45 \times 10^{-9}$ W.

The linewidth of the source was not characterized but it is inferred that it was $\sim$100 MHz. In this experiment, a single beam was used and the cooling relied on collisional mixing of the degrees of freedom. Since this work, no other optical cooling experiment of hydrogen has been reported. Antihydrogen could in principle be cooled using the same technique but this is experimentally more challenging. First of all, the density of antihydrogen in the ALPHA trap is very low (approximately one atom in the trap per experimental cycle) and the atoms have a higher kinetic energy than the hydrogen atoms of Walraven’s experiment. In addition, due to experimental requirements for the creation and trapping of antihydrogen, the ALPHA trap only allows for laser access on one axis. These additional constraints impose higher power requirements on the light source. A recent calculation indicates that it would be possible to cool down the antihydrogen in the ALPHA trap to $\sim$ 20 mK with a pulsed Lyman-α source [42]. The laser cooling simulations were performed assuming a pulsed Lyman-α source with 0.1 $\mu$J of energy per pulse, 100 MHz bandwidth, 10 nsec temporal width and 10 Hz repetition rate. This indicates that the required power level of the laser is 1.0 $\mu$W (in a 2 cm diameter beam) or $6.2 \times 10^{11}$ photons / pulse. As mentioned before, the current ALPHA trap has laser access along one axis (longitudinal z-axis), allowing only 1D Doppler cooling. However, 3D cooling of antihydrogen would be possible by controlling the dynamic coupling between the $x$ – $y$ and $z$ degrees of freedom in the magnetic trap. The coupling can be enhanced by the use of non-harmonic magnetic fields in both $x$ – $y$ and $z$ directions. The current ALPHA trap has five solenoid coils along the $z$ axis to produce the nonlinearity. The trap configuration and the characterization of the solenoids is described in Appendix A. In principle, 3D cooling of the magnetically trapped antihydrogen in the ALPHA trap is possible with the current technology provided the required laser light is available. This is the motivation for undertaking
1.1. Motivation for cooling antihydrogen

the developing of a hydrogen Lyman-α source. The cooling calculation [42] is the basis of the source design discussed in this dissertation. Although some of the specific laser parameters used in the simulation to accomplish the cooling can be changed, and will most likely be refined in the future, as an initial step the source power requirement needs to be realized. That is the focus of our source implementation.

1.1.3 Gravitational interaction between matter and antimatter

Another important experiment that requires laser cooled antihydrogen is the measurement of the gravitational interaction of matter-antimatter systems. In addition to CPT violation, quantum field gravity theories also predict that the gravitational acceleration of matter could be different from that of antimatter [43]. In string theory, for instance, the origin of the asymmetry can be tracked down to the lack of point-like interactions, which violate Lorentz symmetry and by extension, what is known as Einstein equivalence principle (EEP) [44]. EEP is the basis of general relativity. EEP is Einstein’s extension of what is known as the weak equivalence principle (WEP) and states that all the laws of special relativity hold locally in inertial frames. WEP states that freely falling bodies of the same mass and mass distribution should follow the same trajectory [45]. EEP stipulates the gravity interaction of matter-matter and matter-antimatter should be identical. A violation of this symmetry will not necessarily be a proof of any quantum field theory of gravity or even be a proof of CPT violation, as discussed by Nieto et al [38]. It would, however, seriously reduce our confidence in the theory of general relativity, and that is perhaps the main motivation to perform this experiment. As discussed by Roberts et al [46] the evidence of and acceleration on the expansion of the universe points out that either the composition of the universe or the theory describing the particle interactions needs to be revisited. Antihydrogen, given its charge neutrality, is a more suitable candidate for antimatter-matter gravity tests than charged antiparticles such as antiprotons or positrons. In fact the ALPHA collaboration has accepted a detailed proposal for gravity interferom-
1.1. Motivation for cooling antihydrogen

etry measurements [47]. Light-pulse matter wave interferometers split and recombine matter waves using pulses of light. In the interim (between the pulses) the matter waves can follow different paths. During the recombination, matter waves can interfere constructively or destructively depending on their phase. This is reflected on the probability of finding the atom. The phase of the matter waves is shifted during the propagation due to its interactions with different fields. By designing the environment of the matter waves path it is possible to measure the interaction with very high accuracy. Standing waves of nearly resonant laser light are usually used as diffraction gratings in these interferometers. However, in the proposal for antihydrogen gravity measurement at ALPHA the authors acknowledged the difficulty in using these lasers. The problem is that developing a hydrogen Lyman-α source that provides the required power is not an easy task. In fact, currently there are no cw or pulsed Lyman-α sources powerful enough [48]. In their design they use a far-detuned high-energy pulsed laser [47]. A requirement of this scheme is, however, that the antihydrogen is laser cooled to about 20 mK (equivalent to a thermal velocity of approximately 10 m/s) with a pulsed Lyman-α source. The laser cooling strategy is identical to the one discussed in the previous section (pseudo-3D cooling with one laser beam only). Using this temperature they have estimated that the relative precision of the experiment, given various factors such as: the trap magnetic field variations, retroreflective mirror vibration, sensitivity of detectors, background particle noise and number of available atoms, to be $10^{-2}$. This estimation was performed with 250 detected atoms / month. However, upgrades at CERN are expected to increase the availability of atoms to $3 \times 10^3$ atoms / month increasing the relative precision to $2 \times 10^{-6}$. Therefore, the development of a Lyman-α source system with a power level of 1.0 $\mu$W is key for the ongoing ALPHA experiments, which as mentioned before is the motivation of the present work.
1.2 Lyman-\(\alpha\) radiation and hydrogen

Before moving into the details of the source design, some details of hydrogen Lyman-\(\alpha\) radiation and the hydrogen atom are discussed. The emission spectrum of hydrogen is divided into spectral series given by the empirical Rydberg formula.

\[
\frac{1}{\lambda} = R_H^\infty \left( \frac{1}{n^2} - \frac{1}{n'^2} \right) \quad R_H^\infty = 1.0968 \times 10^7 \text{ m}^{-1}
\]

The hydrogen Lyman-\(\alpha\) line is the first emission line in the series resulting from an electron transitioning from the excited states \(n' > 1\) to the ground state \(n = 1\) where \(n\) is the principal quantum number. The series extends from the Lyman-\(\alpha\) line at 121.56 nm for \(n' = 2\) to 91.2 nm for \(n' = \infty\). The discovery of the series by Theodore Lyman between 1906 and 1916 was an important experimental validation of the predictions by the Rydberg formula and Bohr’s model of the hydrogen atom. It is also marks the beginning of the exploration of the extreme ultraviolet (EUV) band of the spectrum.

Lyman-\(\alpha\) radiation is usually considered the upper limit in wavelength of the EUV that extends down to 10 nm, the x-ray threshold. Photons with higher energy than Lyman-\(\alpha\) radiation (> 10.2 eV) are considered ionizing radiation. Radiation below 200 nm is absorbed by air and propagation only occurs in vacuum, hence this part of the spectrum down to the x-ray threshold is called vacuum ultraviolet (VUV). In addition to EUV and VUV, the part of the spectrum from about 150 nm to 30 nm is usually refereed as the XUV region and is characterized by the interaction of photons with the chemical valence electrons of matter. Higher energy photons interact mainly with inner shell electrons and nuclei.

The discovery of the VUV can be attributed to Viktor Schumann \[49,50\] who made three very important contributions (some of which we still use today for the work in this dissertation). First, he used fluorite optics instead of glass, which allowed him to record 182 nm. Second, he developed a special photographic plate without the commonly used gelatine \[51\], which he showed to be a strong UV absorber, and increased the detector sen-
1.2. *Lyman-α radiation and hydrogen*

Sensitivity into the XUV. Third, he constructed vacuum spectrographs and showed that air absorbs wavelength below 200 nm \[50\]. These techniques allowed him to record the spectrum in the region from 185 nm to 123 nm, the Schumann Region. From 1904 to 1906 Theodore Lyman extended the spectrum beyond the Schumann region to 103 nm \[52\]. This was possible by a significant change in instrumentation. He changed transmitting optics to reflective optics. Instead of using a fluorite prism he used a concave ruled grating. He also established the limit of transparency of fluorite to be 126 nm \[53\]. Measurements of wavelength were made and checked by two methods. One was the method of two slits, devised by Lyman, by which the spectrum could be displaced a known distance by utilizing a second slit some distance from the first. The second method was by comparing the second-order spectrum of known lines of longer wavelength with the first-order spectrum of the unknown ultraviolet lines. Using these methods he was able to calibrate and extend the hydrogen spectrum to 95 nm with 0.1 Å resolution. By 1916 he had discovered the series of hydrogen lines now bearing his name. The light in Lymans experiment was generated electrically in a discharge tube of quartz provided with tungsten electrodes and the lines matched the ones predicted by Rydberg in 1890.

The hydrogen Lyman-α line as first measured by Lyman was 121.6 nm (2,460,000 GHz) \[53\]. Since then several corrections to the theory and improvements in experimental techniques have increased the accuracy of the line. We now know that the line actually splits into multiple lines due to the fine and hyperfine structure. However, the fine structure doublet, with the 2p level splitting into \( j = 1/2 \) and \( j = 3/2 \) as seen in Fig. [1.1] is what is referred as the Lyman-α line. This separation is what the best astronomical observations can discern due to the line broadening caused by the particular physical processes in solar and stellar atmospheres. The transition separation is, according to the most up to date accepted experimental measurements, 10,969.05 MHz \[40\]. It can be used to infer astronomical processes and to calculate the amount of hydrogen isotopes (deuterium and tritium) on plasmas. In general the Lyman-α energy value is represented by a single number. The center-of-gravity energy is the average of the fine
1.3. Dissertation overview

structure components weighted by their statistical weights \((2j + 1)\). For the \(2p\) level \(E_{cg}\) is 82,259.11089 cm\(^{-1}\) [40]. The wavenumber of the Lyman-\(\alpha\) line derived from experimental fine structure intervals weighted by calculated line strengths is 82,259.16(14) cm\(^{-1}\) or 2,466,068(3) GHz [40]. This is what we refer to as Lyman-\(\alpha\) line in this work \(\lambda_{Ly-\alpha} = 121.56698\) nm.

1.3 Dissertation overview

This dissertation describes the design, implementation and characterization of a coherent Lyman-\(\alpha\) source prototype (some times refereed as laser in this dissertation) for cooling antihydrogen. The main purpose of this introductory Chapter, besides providing some general background, was to describe the motivation behind this work. The key point is that laser cooling of the antihydrogen trapped in the ALPHA apparatus is essential for the realization of high precision experiments such as CPT violation and antimatter-matter gravity interaction measurements. The remainder of this dissertation moves away from antimatter and fundamental physics research and concentrates on the project objective (i.e. making the source).

The first section of Chapter 2 describes, in detail, the theory of nonlinear optics. Although, there are several books and reviews on the field, the hope is that the chapter is self-sufficient. The specialist can probably skip this section. However, the emphasis is on the theory aspects used in practice for this specific project. The chapter’s remaining sections describe the design and optimization of the source. Chapter 3 is concerned with the implementation and power measurements of the prototype source. It is important to realize that this source prototype implementation is the first step towards a much larger and complex project. Integration of the source with the experimental apparatus at CERN is, perhaps, more technically challenging than this prototype development. It is desirable to have the most robust and reliable source possible before initiating integration. With this in mind, a second source was implemented for comparison. This is briefly described in Chapter 3, as well, along with the evaluation of both sources. Experimental confirmation that the source is resonant with the
Lyman-α transition of hydrogen is described in Chapters 3 and 4.

Spectroscopy of a supersonically expanded beam of hydrogen was performed to confirm that the Lyman-α source is able to induce resonant transitions in cold hydrogen. This experiment required the implementation of the hydrogen beam and its characterization. Chapter 4 describes this development. Some of the highlights of that chapter are the introduction of a "Zeeman bender" as a hydrogen filter and a rubidium-MOT as an atomic beam density sensor. The hydrogen bender uses magnetic field gradients and Zeeman shifts to manipulate the hydrogen beam, and the Rb-MOT uses the elastic scattering cross section of the Rb-beam atoms as an absolute beam flux meter.

The final chapter describes the current work taking place at CERN and the source post-prototype development. In addition, the experiment taking place with the hydrogen beam and Rb-MOT is described along with some of the attempts done to produce an alternative hydrogen source. This chapter is finalized with a summary and conclusion.
Chapter 2

Coherent hydrogen Lyman-\(\alpha\) source design

In the previous chapter I discussed the scientific motivation to develop a hydrogen Lyman-\(\alpha\) source capable of optically cooling trapped antihydrogen. The minimum requirements for such a source were given based on realistic simulations and on the constraints imposed by a technologically achievable experiment in the near term. The most important requirement is power, which is estimated to be 1.0 \(\mu\)W (in a 2 cm diameter beam). In addition pulse duration, frequency and linewidth were specified for a particular cooling scheme. The linewidth used in the simulation was 100 MHz. In addition to these requirements there are several technical specifications that need to be considered. Among these, the most important are the size and the stability of the source system. The system needs to fit into an ionizing radiation protected area of the antimatter experiment and operate for several hours without needing direct human intervention (needs to be stable). The antiproton decelerator, where the ALPHA experiment is located, receives relativistic antiproton beams that cause gamma rays during collisions. The area where the antihydrogen trap is located is a radiation controlled area and personal and equipment need to be protected. The source system can be placed in a protected area approximately 5 meters away from the trap and across a concrete wall. This area, however, can only accommodate human operators for short periods of time. Operators are located approximately 10 meters away from the source in a different compartment. This is the reason the system needs to be compact and very stable.

Coherent Lyman-\(\alpha\) radiation has been generated previously and Table
Chapter 2. Coherent hydrogen Lyman-\(\alpha\) source design

2.1 shows a list of some sources that have been reported. In addition, Lyman-\(\alpha\) can be easily generated with a free electron laser. These devices can produce coherent photons of tuneable energy by accelerating relativistic free electron beams. Furthermore, a continuous wave (cw) Lyman-\(\alpha\) source have been developed and is, in theory, available for an antihydrogen experiment. However, these sources do not meet one or several of the experimental requirements. Free electron lasers are highly complex machines that require large facilities called synchrotrons. Synchrotrons around the world can currently generate a maximum flux of about \(\sim 10^{12}\) photons/sec at the Lyman-\(\alpha\) spectrum. This corresponds to a power of approximately \(2 \mu W\) which according to simulations [42] would be sufficient for cooling antihydrogen in the ALPHA trap. However, synchrotron radiation linewidths are extremely broad (\(\sim 10^{-3}\) eV) for laser cooling. But even for the sole purpose of detecting antihydrogen, for which the power available from synchrotron radiation will certainly suffice and their linewidth is not a limitation, CERN is not equipped with a synchrotron light source facility. There are reports of high power compact free electron laser developments for medical and defence purposes but they are not available [54]. The cw Lyman-\(\alpha\) source developed by Walz et al [55, 56] is perhaps the most advanced source available for laboratory use at the moment. It has been developed with the purpose of cooling antihydrogen and it has several advantages over a pulsed source. However, this system is extremely complex requiring several lasers. Implementing this laser in the form that it has been reported will require more space that is currently available and it is not clear that it could be developed to operate in an unmanned form. Furthermore, there are doubts that the power reported will be sufficient for the experiments envisioned by ALPHA. Finally, a high power source is currently under development at RIKEN-RAL muon facility with the purpose of measuring the anomalous magnetic moment of the muon. The source uses the same nonlinear process used on a tuneable source implemented by Marangos et al [57] to produce low energy polarized muons.

Besides synchrotron radiation, any other techniques for producing coherent hydrogen Lyman-\(\alpha\) radiation requires the use of nonlinear processes
in gases. In this chapter, I describe the design and optimization of a coherent light source based on nonlinear frequency conversion. The development of the source was approached in steps of increasing complexity. The first step was to develop a source able to detect hydrogen (and antihydrogen) and that generated, at least, the power specified for cooling. The idea is to develop a system that can be used for initial experiments and also be updated as to increasingly meet the experimental requirements such as linewidth and tuneability. With regards to the actual implementation of the source, based on a literature review, the most common choice seems to be a third harmonic generation (THG) process in a noble gas. Several sources have been developed this way \[59-62\] and there is plenty of information about it. However, it was decided to develop a system which uses a different nonlinear process. The specific design used is very promising with regards to the potential power, linewidth and tuneability. The THG was still implemented in parallel to this development with the purpose of having a second source for comparison. This would allow us to evaluate the best system to continue into the next step of integration with the ALPHA apparatus.

The chapter has three sections. First, the background theory of non-
2.1 Nonlinear optics

Nonlinear optics necessary to understand how the Lyman-α source works is explained. Central to this discussion are the concepts of the nonlinear susceptibility and phase matching. This is followed by a description of the design and optimization of the source. The source prototype implementation and characterization are delayed until the next chapter where the different detection techniques used are discussed. As part of the characterization, spectroscopy of a supersonically expanded hydrogen beam was performed. Details of the hydrogen beam implementation, characterization, and the spectroscopy are discussed in Chapter 4.

2.1 Nonlinear optics

Nonlinear optics processes such as frequency sum and difference (frequency mixing) are commonly used to extend the spectral range of lasers [67]. This is mostly done using crystals and visible and infrared light is commonly generated. Crystals, however, absorb radiation in the vacuum ultraviolet spectrum (VUV) range (200 nm-100 nm). The onset of saturation, for instance, of β-barium borate “BBO” crystals is 189 nm [68]. For this reason, frequency mixing in the VUV spectrum range is usually done in gases. In the following discussion we explore a particular process known as four-wave mixing (FWM) which is the most commonly used process for VUV light generation.

FWM arises from the interaction of four coherent optical fields through the third order nonlinear susceptibility. This phenomenon includes many diverse processes such as third-harmonic generation, stimulated Raman scattering, and Raman induced Kerr effects. Central to the discussion is the third order nonlinear susceptibility $\chi^{(3)}$ which is ultimately responsible for all FWM processes $^2$.

Some of the early works, with New and Ward [69] being perhaps the first one on UV light generation by nonlinear processes in gases (generated

$^2$In fact, the reason FWM ($\chi^{(3)}$) is used to generate VUV is that gases, which are transparent to VUV, do not show second order nonlinear susceptibility ($\chi^{(2)}$) due to symmetry reasons
231.4 nm by third harmonic generation), reported very low conversion efficiencies. It was not until the work of Harris and Miles [70], and Young et al [71], showed that conversion efficiency could be increased by several orders of magnitude by using a two-component system that the technique became of practical interest. In this scheme one component is used to provide the overall refractive index and the second component provides the nonlinear susceptibility for the particular process. With two components, phase matching is possible, generating conversion efficiencies of \( \sim 10^{-7} - 10^{-6} \) [6]. The final advance that made the technique useful for practical applications was the demonstrations by Bloom et al, Hogdson et al and Leung et al [72–74] of resonant enhancement of the nonlinear susceptibility. This technique increases the conversion efficiency by several orders of magnitude (\( \geq 10^{-4} \)) [67, 68] by using a two-photon resonance transition. Much effort to develop new mixing schemes has been overtaken since then with the goal of increased conversion efficiency and greater tuning range extending well into the extreme ultraviolet spectrum (XUV) (100 nm -10 nm). For instance doubly-enhanced sum-frequency mixing have been demonstrated by Softley et al, where in addition to the two-photon resonance a second resonance at the sum-frequency is exploited [75, 76]. There are published reviews of the subject with Vidals [77] being perhaps the most comprehensive one.

A very commonly used mixing technique uses noble gases (Xe, Kr, Ar, and Ne) as the nonlinear media. Noble gases have an appropriate energy-level structure and are convenient to work with. Metal vapours such as Hg are also suitable for VUV and XUV generation but are not as convenient to work with and require heat pipes to maintain the metal in a vapour state. An additional advantage of using atomic gases is their inherent narrow linewidths which allow for the generation of very narrow linewidth coherent light (<1 cm\(^{-1}\)) [78].

In practical terms for radiation generated between 200 nm and 120 nm there are windows reasonably transparent (MgF\(_2\) and LiF) and vacuum-sealed cells filled with the mixing gases can be used as the nonlinear medium and light source. Since oxygen is the main absorber of radiation in this
2.1. Nonlinear optics

wavelength range, vessels filled with argon can be used as the working medium (for VUV processes that do not require vacuum). If shorter wavelength (≤ 120 nm) radiation is required the light source cell has to be connected directly to a vacuum chamber (not possible to use a LiF window with cut off at 105 nm) via a differential pumping scheme. In this case the nonlinear gas is usually injected into the vacuum in the source cell with a pulsed valve and nozzle system [79-82]. In most of these schemes, the fundamental laser beams are focused into the nonlinear medium in order to increase the field strength and improve the nonlinear coupling.

The most common FWM processes used to generate VUV and XUV are [83]:

- Non-resonant tripling:
  \[ \omega_{\text{VUV}} = 3\omega_1. \]

- Resonant Sum Frequency Mixing:
  \[ \omega_{\text{VUV}} = \omega_1 + \omega_2 + \omega_3 \text{ (or } 2\omega_1 + \omega_2). \]

- Resonant Sum-Difference Frequency Mixing:
  \[ \omega_{\text{VUV}} = \omega_1 + \omega_2 - \omega_3 \text{ (or } 2\omega_1 - \omega_2). \]

- Anti-Stokes stimulated Raman scattering:
  \[ \omega_{\text{VUV}} = \omega_1 - \omega_2 + \omega_3. \]

The most convenient mixing scheme for producing a widely tuneable output is sum-difference. The reason is there are no restrictions on the sign of the wave-vector mismatch “Δk” [84] (this point is further explained below). Non-resonant tripling is convenient as only one primary beam is needed, greatly reducing the complexity of the system. We will concentrate on these two processes.

The following discussion of nonlinear optics is based on a perturbative approach first introduced by Bloembergen [85]. In this approach, the nonlinear optical properties of the material are fully described by the nonlinear susceptibilities, which are obtained by applying the Fourier transform to the nonlinear Green’s function. The nonlinear Green’s function can be
obtained using a generalization of Kubo’s linear response theory [86]. The perturbative theory is generally in good agreement with the experimental data, and comparisons have been published by Butcher [87] and Boyd [88]. This approach is a semiclassical analogue of Feynman diagrams formalism [89] and diagrammatic techniques to calculate the perturbation terms of the density matrix have been suggested by Yee et al [90].

2.1.1 Nonlinear susceptibility

The interactions of electromagnetic waves with matter can be described by Maxwell’s equations. At optical frequencies the materials of interest are usually non-magnetic. Also, as long as plasmas are not involved, we can assume that the mediums are source and current free. Under these constraints on the Maxwell’s equations it is possible to derive the wave equation.

\[
\nabla^2 \vec{E} + 4\pi \nabla (\nabla \cdot \vec{P}) - \frac{1}{c^2} \frac{\partial}{\partial t} \vec{E} = \frac{4\pi}{c^2} \frac{\partial}{\partial t} \vec{P},
\]

(2.1)

with

\[
\rho_b = - (\nabla \cdot \vec{P}),
\]

(2.2)

and \(\rho_b\) is the charge density attributable to bound charges (i.e. charges arising from the polarization of neutral atoms). The term \(4\pi \nabla (\nabla \cdot \vec{P})\) is often dropped in the literature, which is usually correct, but it should be considered in nanoscale, meta-materials and interface calculations.

The response of the medium to an electromagnetic perturbation is fully described by the above equation. The polarization term, \(P(\vec{r}, t)\), provides the full description of the light-matter interaction and that is what we will now derive. The polarization can be expressed by the following convolution:

\[
P_i^{(T)}(\vec{r}, t) = \iiint_{-\infty}^{\infty} G_{ij}^{(T)}(\vec{r}, t'; \vec{r} - \vec{r}', t' - t) E_j^{loc}(\vec{r}', t') dt' d\vec{r}'.
\]

(2.3)

The electric field, \(E^{loc}(\vec{r}, t)\), is the local perturbation (at \(\vec{r}\) and \(t\)) caused by an external macroscopic field and \(G(\vec{r}, t)\) is the linear Green’s function accounting for the response of the system. In general \(E^{loc}(\vec{r}, t)\) is not the
2.1. Nonlinear optics

macroscopic applied field. However, it is customary to carry on with the derivation assuming $E^{loc}(\vec{r},t) = E(\vec{r},t)$ and modifying the final result by local field corrections [91–93].

The convolution form of the polarization is particularly advantageous for calculation as it can be reduced to a product of Fourier transforms (in space and time).

$$P_1(\vec{k}, w) = f \{ \mathcal{F}[P_1(\vec{r}, t)] \} = f \{ \mathcal{F}[G_{ij}(\vec{r}, t)\mathcal{F}[E_j(\vec{r}, t)]] \} = f \{ \chi_{ij}(\vec{k}, w)E_j(\vec{k}, w) \}.$$  \hspace{1cm} (2.4)

The susceptibility, $\chi$, is a tensor that contains all the information of the system. Additionally, in the optical frequency range the susceptibility of the system does not depend on the magnitude of the wave vector, which obeys the condition $|\vec{k}|d \ll 1$ where $d$ is the characteristic order of dimensions of the constituent elements of the material. For the atomic gases considered here $d$ is on the order of the Bohr radius. This is called the dipole approximation and is equivalent to dropping all the $\vec{r}$ dependences on the convolution relation [87].

$$\chi^{(n)}_{i_1...i_n}(w) \equiv \lim_{\vec{k} \to 0} \chi^{(n)}_{i_1...i_n}(\vec{k}, w).$$ \hspace{1cm} (2.5)

To calculate the nonlinear terms of interest we can generalize the definition of polarization. The total polarization can then be expressed as the sum of linear and nonlinear contributions at each order of nonlinearity:

$$\vec{P}(t) = \vec{P}^{(1)}(t) + \vec{P}^{NL}(t),$$ \hspace{1cm} (2.6)

$$\vec{P}^{NL}(t) = \sum_{n=2}^{\infty} \vec{P}^{(n)}(t).$$ \hspace{1cm} (2.7)

And the n-order nonlinear polarization can be defined as a multiple convolution product:

$$P^{(n)}_i(t) = \int_{-\infty}^{\infty} G_{i_1...i_n}^{(n)}(t\cdots t_n)E_{j_1}(t-t_1)\cdots E_{j_n}(t-t_n)dt_1\cdots dt_n.$$ \hspace{1cm} (2.8)
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As before, we compute the Fourier transform and obtain an expression for the nonlinear susceptibility.

\[
P_i^{(n)}(\omega) = \int_{-\infty}^{\infty} \chi_{i,j_1,\cdots,j_n}^{(n)} \left[ \sum_{l=1}^{n} \omega_l \cdot \omega_1, \cdots, \omega_n \right] E_j(\omega_1) \cdots E_{j_n}(\omega_n) \\
\times \delta \left( \omega - \left( \sum_{l=1}^{n} \omega_l \right) \right) d\omega_1 \cdots d\omega_n, \quad (2.9)
\]

with

\[
\chi_{i,j_1,\cdots,j_n}^{(n)} \left[ \sum_{l=1}^{n} \omega_l \cdot \omega_1, \cdots, \omega_n \right] = \int_{-\infty}^{\infty} G_{i,j_1,\cdots,j_n}^{(n)}(t_1, \cdots, t_n) \exp \left[ \sum_{l=1}^{n} \omega_l t_l \right] dt_1 \cdots dt_n,
\]

(2.10)

The Dirac \( \delta \)-function guarantees that the sum of the arguments of the Fourier transforms of the electric field equals the argument of the Fourier transform of the polarization. Physically, each \( \omega \) component of the polarization results from the interaction of \( n \) photons mediated by the material susceptibility function.

2.1.2 Quantum-mechanical expression of the nonlinear Green’s function

The susceptibility is ultimately a function of the microscopic interactions of the material. Therefore, we would like to derive a quantum mechanical expression for the nonlinear Green’s function. The quantum mechanical description that we follow here is based on the principle of “minimal coupling” approximation. In general terms this refers to the coupling of fields that involve only the charge distribution. Minimal coupling disregards higher multipole moments and magnetic moments of the particles. Under the approximation, the Schrödinger equation for a single particle takes the form:

\[
H = \frac{1}{2m} \left( -i\hbar \nabla - q\vec{A}(\vec{x}, t) \right)^2 + q\phi(\vec{x}, t),
\]

(2.11)
2.1. Nonlinear optics

and

\[ i\hbar \partial_t \psi(\vec{x}, t) = H \psi(\vec{x}, t), \quad (2.12) \]

where \( \vec{A} \) and \( \psi \) are the vector and scalar potentials and \( q \) is the charge of the particle. For a many body system under the dipole approximation and using a Legendre transformation we arrive at a total Hamiltonian:

\[ H = H_0 + H_1, \quad (2.13) \]

with

\[ H_0 = T + V + H_{ee} = \sum_\alpha V(\vec{r}_\alpha) + \sum_{\alpha \neq \beta} \frac{e^2}{|\vec{r}_\alpha - \vec{r}_\beta|}, \quad (2.14) \]

and

\[ H_1 = e \sum_\alpha \vec{r}_\alpha \cdot \vec{E}(t), \quad (2.15) \]

where the charge of the electron has been replaced \( q = -e \). Here, all the time dependence is in the interaction part, usually referred as the Hamiltonian in the gauge of length.

As we are dealing with ensembles of particles and are interested in the statistical properties it is convenient to work with the density operator. Calculations can also be performed using the perturbed particle wavefunctions. However, introduction of damping due to dephasing and population decay is more straightforward using the density matrix approach

\[ \rho = \sum_{a,b} \rho_{ab} |a\rangle \langle b|. \quad (2.16) \]

The macroscopic polarization per unit volume can be defined as the expectation value of the dipole moment per unit volume where the initial condition is given by the Boltzmann equilibrium distribution [85-87].

\[ P_i^{(n)}(t) \equiv \frac{1}{V} Tr \left\{ -e \sum_{\alpha=1}^{N} \vec{r}_\alpha \rho^{\alpha}(t) \right\}, \quad (2.17) \]
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with

\[
\rho(0) = \frac{\sum a \exp -\frac{E_a}{k_B T} |a\rangle\langle b|}{\sum a \exp -\frac{E_a}{k_B T}}. \tag{2.18}
\]

The density operator obeys the Liouville differential equation (with relaxation terms ignored).

\[
i\partial_t \rho(t) = [H, \rho(t)] = [H_o, \rho(t)] + [H_I, \rho(t)]. \tag{2.19}
\]

This equation can be solved using perturbation theory techniques by expanding the density operator in a series that leads to a system of coupled differential equations:

\[
\begin{align*}
i\partial_t \rho^{(1)}(t) &= [H_o, \rho^{(1)}(t)] + [H_I, \rho(0)] \\
i\partial_t \rho^{(2)}(t) &= [H_o, \rho^{(2)}(t)] + [H_I, \rho^{(1)}(t)] \\
& \downarrow \cdots \downarrow \\
i\partial_t \rho^{(n)}(t) &= [H_o, \rho^{(n)}(t)] + [H_I, \rho^{(n-1)}(t)].
\end{align*}
\]  

\(\tag{2.20}\)

It is convenient to use the interaction representation for the evolution of the position operators

\[
\begin{align*}
r_i^a(-t) &= \exp \left[ \frac{iH_o(-t)}{\hbar} \right] r_i^a \exp \left[ -\frac{iH_o(-t)}{\hbar} \right]. \tag{2.21}
\end{align*}
\]

Considering the n-differential equation in the system, and with the boundary condition \(\rho^{(n)}(0) = 0\), the solution of this equation can be obtained recursively by solving each increasing perturbation order \([85, 86]\). At the end \(\rho^{(n)}(t)\) can be expressed as a function of the dipole operators and of
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\( \rho (0) \) only:

\[
\rho^{(n)}(t) = \left( \frac{e^{i\hbar}}{-i\hbar} \right)^n \int_{-\infty}^t \cdots \int_{-\infty}^{t_{n-1}} E_{j_1}(t_1) \cdots E_{j_n}(t_n) \,* \\
\left[ \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_{j_1}(t_1 - t) \cdots \sum_{\alpha = 1}^N r^{\alpha}_{j_n}(t_n - t), \rho(0) \right] \cdots \right] \, dt_1 \cdots dt_n. \quad (2.22)
\]

Using the following relation:

\[
Tr \left\{ \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_i(-t_1), \cdots, \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_i(-t_n), \rho(0) \right\} = \\
(-1)^n \left\{ \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_i(-t_1), \cdots, \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_i(-t_n), \rho(0) \right\}, \quad (2.23)
\]

and the definitions of polarization:

- expectation value of the dipole operator per unit volume
- convolution of the nonlinear Green’s function times \( n \) electric fields

it is possible to derive the following relation:

\[
G^{(n)}_{i_{j_1} \cdots j_n}(t_1, \cdots, t_n) = \frac{e^{n+1}}{V(-i\hbar)^n} \int_{-\infty}^{\infty} \Theta(t_1) \cdots \Theta(t_n - t_{n-1}) \,* \\
Tr \left\{ \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_{j_1}(-t_1), \cdots, \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_{j_n}(-t_n), \sum_{\alpha = 1}^N \sum_{\alpha = 1}^N r^{\alpha}_i \right\} \rho(0) \right\}. \quad (2.24)
\]

Finally, with the definition of the nonlinear Green’s function we can express
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the n-order nonlinear susceptibility of a general quantum system.

\[ \chi^{(n)}_{i,j_1\cdots j_n} \left( \sum_{l=1}^{n} \alpha_i \omega_1, \cdots, \omega_n \right) = \]

\[ \int_{-\infty}^{\infty} G_{i,j_1\cdots j_n}^{(n)}(t_1, \cdots, t_n) \exp \left\{ \sum_{l=1}^{n} i \omega_l t_l \right\} dt_1 \cdots dt_n = \]

\[ - \frac{e^{n+1}}{\sqrt{(-i\hbar)^n}} \int_{-\infty}^{\infty} \Theta(t_1) \cdots \Theta(t_n - t_{n-1})^* \]

\[ \text{Tr} \left\{ \left[ \sum_{\alpha=1}^{N} r_{jn}^\alpha (-t_n), \cdots, \left[ \sum_{\alpha=1}^{N} r_{j_1}^\alpha (-t_1), \sum_{\alpha=1}^{N} r_i^\alpha \right] \cdots \rho(0) \right] \exp \left\{ \sum_{l=1}^{n} i \omega_l t_l \right\} dt_1 \cdots dt_n, \]

(2.25)

In the preceding derivation relaxation terms were neglected but can be included in the equation of motion:

\[ i \partial_t \rho(t) + \left( \frac{i \hbar}{\Gamma} \right) \rho(t) = [H_o, \rho(t)] + [H_I, \rho(t)], \]

(2.26)

where \( \Gamma \) the damping constant includes the transverse (off-diagonal) and longitudinal relaxation terms. A formal solution of the components \( \rho^{(i)}(t) \) at frequency \( \pm \omega \) has been worked out by Pell and Vidal [94] in terms of the iterated integral:

\[ \rho_{ij}^{(n)}(t) = \frac{i}{\hbar} \exp \left\{ -i \omega_{ij} t \right\} \int_{-\infty}^{t} A(t') \left[ \exp \left\{ -i (\omega - \omega_{ij}) t' \right\} + \exp \left\{ -i (\omega + \omega_{ij}) t' \right\} \right] \\
\times \left[ r_{ij}, \rho^{(n-1)}(t') \right] dt', \]

(2.27)

where \( \omega_{ij} = (E_i - E_j)/\hbar - i/\Gamma_{ij} \) is the energy difference (eigenfrequencies) between level \( i \) and \( j \). The complex term \( i/\Gamma \) is important in cases where we operate close to resonances.

Using Eq. (2.25) it is possible to derive explicit expressions for the non-linear susceptibility of specific processes in media with discrete quantum levels. This is done by performing the Fourier transform integral of the
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Green’s function and expanding the time-dependent position operators \( r \). Of particular interest for this work is the third order nonlinear susceptibility of FWM processes and, specifically, non-resonant third order susceptibility and resonant frequency sum-difference. Formulas for the FWM processes \( \omega_4 = \omega_1 + \omega_2 + \omega_3 \) which are far away from resonances have been given by Armstrong et al [95]. For situations where damping constants need to be included (resonant FWM) formulas have been worked out by Bloemebergen and Shen [96].

For FWM sum-difference interaction in a gas system the susceptibility per atom is (with \( r_{ij} \) in Bohr radius units and \( \omega \) in cm\(^{-1} \) which is customarily used for calculation):

\[
\chi^{(3)}(-\omega_4; \omega_1, \omega_1, \pm \omega_2) = \frac{1}{3} \frac{(ea_o)^4}{(hc)^2} \sum (r_{0i}r_{ij}r_{jk}r_{k0})
\]

\[
\times \left[ \frac{1}{(\omega_{j0} + \omega_2)(\omega_{j0} - (\omega_1 \pm \omega_2))} + \frac{1}{(\omega_{i0} - \omega_1)(\omega_{j0} - (\omega_1 \pm \omega_2))} \right]
\times \left[ \frac{1}{\omega_{k0} - \omega_4} + \frac{1}{\omega_{k0} + \omega_1} \right]
\]

\[
+ \frac{1}{(\omega_{i0} - \omega_1)(\omega_{j0} - 2\omega_1)} \left[ \frac{1}{\omega_{k0} - \omega_4} + \frac{1}{\omega_{k0} \pm \omega_2} \right]
\]

\[
+ \left[ \frac{1}{(\omega_{i0}^* + \omega_2)(\omega_{j0}^* + (\omega_1 \pm \omega_2))} + \frac{1}{(\omega_{i0}^* + \omega_1)(\omega_{j0}^* + (\omega_1 \pm \omega_2))} \right]
\times \left[ \frac{1}{\omega_{k0}^* + \omega_4} + \frac{1}{\omega_{k0} - \omega_1} \right]
\]

\[
+ \frac{1}{(\omega_{i0}^* + \omega_1)(\omega_{j0}^* + 2\omega_1)} \left[ \frac{1}{\omega_{k0}^* + \omega_4} + \frac{1}{\omega_{k0} \pm \omega_2} \right].
\] (2.28)

In the gas systems that we are considering it is possible to write each matrix element as a coefficient times a reduced matrix element using the
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Wigner-Eckart theorem \cite{97}:

\[ r_{ab} = \alpha_{J_a m_a} | r_0 \rangle \beta_{J_b m_b} = (-1)^{J_a - m_a} \left( \begin{array}{cc} J_a & 1 \\ m_a & \Delta m_0 \\ m_b & \end{array} \right) \langle \alpha | \langle r \parallel \beta \rangle . \] (2.29)

The difference on magnetic quantum number \( \Delta m_\theta \) in the \( 3j \)-symbol is \( \pm 1 \) for circularly polarized light and 0 for linearly polarized. The selection rule associated with \( 3j \)-symbol symmetry yields the following condition for the susceptibility numerator:

\[ \left( r_{0i} r_{ij} r_{jk} r_{k0} \right) \Rightarrow \Delta m_{0i} + \Delta m_{ij} + \Delta m_{jk} + \Delta m_{k0} = 0. \] (2.30)

There are two practical implications from these equations that are worth pointing out. First, from the matrix elements expressions it is evident that two right-hand and one left-hand circularly polarized waves produce a left-hand circularly polarized wave. Three right-hand or left-hand circularly polarized waves do not produce a frequency sum \cite{77}. Second, the selection rules indicate that the states \( |0 \rangle \) and \( |j \rangle \) have opposite parity to the \( |i \rangle \) and \( |k \rangle \) states. To enhance the susceptibility the resonant denominator should be minimized (see Eq. (2.28)). Since \( \omega_{k0} \) and \( \omega_{l0} \) are allowed transitions, and \( \omega_{j0} \) is not allowed, the term \( (\omega_{j0} - 2\omega_1) \) should be minimized. In plain language, we wish to operate at a resonance in order to minimize the resonant denominator in the susceptibility, and this is achieved in an optimal way by avoiding the one-photon resonances (which would lead to absorption of the fundamental wave) and by avoiding the three-photon resonance (which would lead to absorption of the produced light. This is the basis of two-photon resonant enhanced FWM and was demonstrated first by Bloom et al, Hodgson et al and Leung et al, \cite{72–74}. This will be revisited in the next section.

Finally, as mentioned at the beginning, the susceptibility derivation used \( E_{loc}(r, t) = E(r, t) \), and the final result needs to be modified by local field corrections. In isotropic media such as gases the depolarization field
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can be accounted for by multiplying each field component by:

\[ L = \frac{\epsilon(\omega) + 2}{3}, \]  

(2.31)

where \( \epsilon(\omega) \) is the linear dielectric constant at \( \omega \) \[85\]. The macroscopic susceptibility is proportional to the number of scattering centers times the local field corrections. In rarified gases \( L \approx 1 \) and the macroscopic susceptibility is:

\[ \chi_T = N \chi_a, \]  

(2.32)

where \( N \) is the density number. If the density is very large, such as in cryogenic crystals of noble gases, local field corrections become important.

2.1.3 Four wave mixing solution

The third order polarization describes a coupling between four waves, each with its own direction of propagation, polarization, and frequency. If this nonlinear polarization is substituted into the nonlinear wave equation, Eq. (2.1), a set of four coupled wave equations may be found for the fields. As long as the variation of the field amplitude is small in an optical period the differential equations can be reduced to first order. This is called the slowly varying envelope approximation and is generally valid for laser radiation. The equations for the field magnitudes can be written in the following form:

\[ \frac{\partial E_j}{\partial z} + n_j \frac{\partial E_j}{c \partial t} = \frac{2\pi i \omega_j}{cn_j} P_{j}^{NL} \exp\{ik_jz\} - \frac{\kappa_j}{2} E_j, \]  

(2.33)

where \( n \) is the refractive index and \( \kappa = (\omega \epsilon_i) / (cn) \) is the absorption coefficient. The third order nonlinear polarization is:

\[ P^{(3)}_s(\omega_s) = \frac{3}{2} N \chi^{(3)}_{s,j_1,j_2,j_3}(\omega_s;\omega_1,\omega_2,\omega_3)E_1 E_2 E_3, \]  

(2.34)
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where \( \omega_s = \omega_4 \) is the generated wave that we are trying to solve the equations for.

These equations cannot be solved in general and the most common approach is to assume:

- plane wave solutions,
- the energy transferred from the input fields to the fourth field is a negligible fraction of the total energy of the fields.

The solution of the intensity in the small signal approximation as first derived by Bey et al \([98]\) is:

\[
\frac{I_s}{n_s} = \left[ \frac{24\pi^2 \omega_s}{c^2 n_s^3} NL \chi^{(3)}_{s1j2j3}(\omega_s;\omega_1\omega_2\omega_3) \right]^2 \frac{I_1^0 I_2^0 I_3^0}{n_1 n_2 n_3} F(\Delta kL, \tau_i \tau_s),
\]

with the optical depth:

\[
\tau_j = k_j L = \sigma_j^{(1)}(\omega_j)NL \quad \tau_s = \tau_1 + \tau_2 + \tau_3
\]

and the phase matching factor:

\[
F(\Delta kL, \tau_i, \tau_s) = \frac{\exp(-\tau_i)\exp(-\tau_s) - 2\exp\left(-\frac{\tau_i + \tau_s}{2}\right)\cos(\Delta kL)}{\left[\left(\frac{\tau_i - \tau_s}{2}\right)^2 + [\Delta kL]^2\right]},
\]

where \( L \) is the total length of the nonlinear medium, \( \sigma \) is the one-photon absorption cross section and \( \Delta k = k_s - k_1 - k_2 - k_3 \).

### 2.1.4 Phase matching factor

The phase matching factor is an expression of conservation of energy and momentum. All the four waves must maintain a constant phase in order to avoid destructive interference. In order to optimize the FWM process one has to minimize the three variables \( \Delta kL, \tau_i \) and \( \tau_s \). The limiting cases of the phase matching factor are \([97]\):
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- Negligible absorption cross sections $\tau_i = 0, \tau_s = 0$:

  \[ F(\Delta kL, \tau_i = 0, \tau_s = 0) = \frac{\sin(\Delta kL/2)^2}{(\Delta kL/2)^2}. \]  

(2.38)

- Large absorption cross section $\tau_i \gg 1, \tau_s \gg 1$:

  \[ F(\Delta kL, \tau_i \gg 1, \tau_s \gg 1) = \frac{1}{[(\tau_s - \tau_i)/2]^2 + (\Delta kL)^2}. \]  

(2.39)

- Phase matched system $\Delta kL = 0$:

  \[ F(\Delta kL = 0, \tau_i, \tau_s) = \left[ \exp(-\tau_i/2) - \exp(-\tau_s/2) \right]^2 / (\tau_s - \tau_i)/2. \]  

(2.40)

Finally for the case of large absorption with $\tau_s > \tau_i$ and $\Delta kL = 0$ we have

\[ \frac{I_s}{n_s} = \left( \frac{48\pi^2 c^2 n_s^2}{s_{3, j_1 j_2 j_3}^{(3)}(\omega_{s}; \omega_{1}, \omega_{2}, \omega_{3}) a_{s}^{(1)}} \right)^2 \frac{I_1^n I_2^n I_3^n}{n_1 n_2 n_3}. \]  

(2.41)

The last case is the most interesting, as it is always possible to find incident optical frequencies away from resonances but the absorption cross section of the fourth wave cannot be neglected [77]. The intensity is not dependent on the density, $NL$, and is limited only by the absorption cross-section and susceptibility.

The validity of the results derived using the plane wave approximation is limited, since the fields used experimentally are Gaussian. In the case of very short interaction length (smaller than the Rayleigh length) the phase fronts are approximately planar and if $\Delta kL \Rightarrow 0$ then the intensity expression for $I_s/n_s$ still applies. However, in general these expressions need to be modified since Gaussian beams have a phase shift, the Gouy phase shift, of $\pi$ at the focus. Assuming a $TEM_{00}$ mode, negligible cross-sections (no absorption), a collinear arrangement of beams, and identical confocal parameter $b = 2kR^2$ where $R$ is the $1/e^2$ radius of the intensity distribution,
2.2. Source design and optimization

the phase matching factor as calculated by Bjorklund \[84\] is:

\[ F(\Delta kL, b/L) = \frac{1}{L^2} \left[ \int_{-L/2}^{L/2} \frac{\exp(-i\Delta kz)}{(1 + i2z/b)^2} dz \right]^2. \]  

(2.42)

The optimal conditions for FWM processes are:

\[ \Delta kb = \begin{cases} 
-2 & \omega_s = \omega_1 + \omega_2 + \omega_3 \\
0 & \omega_s = \omega_1 + \omega_2 - \omega_3 \\
+2 & \omega_s = \omega_1 - \omega_2 - \omega_3.
\end{cases} \]  

(2.43)

Sum-difference frequency mixing with a condition \( \Delta kb = 0 \) is the most forgiving and therefore the preferred process for widely tuneable lasers. In the case of sum frequency (or THG) the phase matching is more restricted. However, for THG the phase matching only requires two wavelengths and is tremendously simplified. Finally, excessive focusing will excite higher order perturbations generating competing processes not included in the previous analysis. According to Vidal \[77, 97\] the optimum power conversion in a THG phase matched system is \( b=L \), experimentally.

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The total harmonic power is obtained by integrating the power density over the beam profile which is usually approximately Gaussian. In FWM processes the power of the generated wave can be expressed as:

\[ P_s \propto N^2 \cdot [\chi^{(3)}]^2 \cdot P_1 \cdot P_2 \cdot P_3 \cdot F. \]  

(2.44)

All of these factors are related and it is not possible to optimize one of them without considering the others. However, the first step is to select a medium and a process. The main criteria is usually to maximize the susceptibility.
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2.2.1 Resonance-enhancement of FWM processes

The susceptibility of the medium ultimately emerges from its electronic structure. Choosing a medium with an appropriate energy structure is perhaps the most important step towards designing and optimizing a source. Important factors are the total coefficient $\Re\{\chi^{(3)}\}$, as it affects the power generated directly, and also the shape of the dispersion as it affects the phase matching factor. In general, a medium with a rapidly changing dispersion $\Re\{\chi^{(1)}(\omega)\}$ (linear susceptibility) is not desirable as phase matching is very difficult. Furthermore, even if phase matching is achieved, the usable bandwidth of the input beams is very limited. The availability of the input lasers is, of course, important as well, and it reduces the choices of available resonances in nonlinear medium. The susceptibility:

$$\chi^{(n)} \propto \frac{\text{dipole moments}}{\text{resonances}}, \quad (2.45)$$

is a function of the oscillator strengths and resonances. Resonant terms in the denominator increase the overall susceptibility. However, not all resonances are good. One-photon resonance occurs when the incident photon frequency matches that of one of the allowed dipole transitions from the ground state. Strong absorption is a problem and as a result the resonant denominator cannot be made too small. Near resonance is advantageous as long as the variation of the dispersion is not too great as to reduce the coherence length to the point where phase matching is impossible. Three-photon resonances, which involves combinations of three photon frequencies, are in principle the same as one-photon resonance. They involve transitions between different parity states and the angular momentum restrictions depend on the particular coupling of the medium ($L - S$ or $J_1 - J_2$). These type of resonances are important for processes that require negative dispersion, such as sum frequency, but they usually lead to absorption as the one-photon case. Two-photon resonances involve combinations of two photon frequencies. They require states with the same parity and $\Delta J = 0, \pm 1, \pm 2$. Two-photon absorption is generally much weaker than the one- or three-photon cases. Fig. 2.1 shows diagrams for the different cases.
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Figure 2.1: FWM processes can be enhanced by the use of resonances. Energy diagrams of the three types of resonances available for FWM are shown for a sum-frequency resonant process. The only type that would not result in absorption of one of the input beams or the generated beam is the two-photon resonant case, because the transition is electric dipole forbidden.

With regards to the medium, alkali vapours have convenient energy levels given the available input laser and K, Rb and Na have been used to generate VUV. Hg is also very commonly used for VUV generation. The difficulty of using these sources relies on the requirement of using heat pipes to maintain the vapour state of the metal. Noble gases are more attractive in this regard. In addition, noble gases have the highest ionization energy of all elements; ionization is always a problem when generating VUV light. Kr for instance has an appropriate energy level and reported high third order nonlinear susceptibilities. Xe is also very appealing gas but the cost / mol of Xe is about two orders of magnitude higher than Kr.
Fig. 2.2 shows a particular resonance-enhanced sum-difference FWM process in Kr. The Grotrian diagram with the relevant energy levels and transitions is shown in Fig. 2.3.

In this scheme the two-photon resonance-enhanced FWM (RE-FWM) uses the resonant condition (see Eq. (2.28)) of \( \omega_{j0} = 2\omega_1 \) with \( \omega_1 = \omega_2 \). For the generation of Lyman-\( \alpha \) radiation at 121.56 nm, Kr gas is an efficient nonlinear medium since the \( 4s^24p^55s^2[1/2]_{1/2} \) \( J \) = 1 level is just \( \sim 1300 \text{ cm}^{-1} \) below the Lyman-\( \alpha \) frequency, which adds another nearly resonant condition of \( \omega_{kg} \sim 2\omega_1 - \omega_3 \) in the third order susceptibility. This process is very promising given the extremely favourable resonance conditions (i.e. two photon resonance and nearly resonance between excited states). In addition, Kr energy states conveniently suppress competing processes such as \( \omega_1 + \omega_3 \) and one-photon resonances (from ground state). Competing processes, such as multiphoton absorption are important saturation mechanisms [75, 76], and are discussed in the next section. The scheme requires a two-photon resonant beam \( \omega_R = \omega_1 = \omega_2 = 2\pi \times 1.4818 \times 10^{15} \text{ Hz} \) or 202.31 nm and a tuneable beam \( \omega_T = \omega_3 = 2\pi \times 4.9753 \times 10^{14} \text{ Hz} \) or 602.56 nm to adjust the output wave \( \omega_{VUV} = 2\omega_R - \omega_T \). Both of these wavelengths are available at sufficient powers.

Using Eq. (2.28) is possible to calculate the nonlinear susceptibility of Kr for this process. The sum is over all exited states connected by allowed dipole transitions (see Fig. 2.3). However, the basic selection rules, which apply only to simple configurations, obey strict \( LS \) coupling. Inert gases, except helium, are more complex and the \( L \) and \( S \) levels interact. Their structure is usually described by \( Jj \) or \( jl \) coupling schemes and both are found in the literature. Inert gases have six electrons in the outermost subshell. These are the electronic structures of argon (argon plays an important role in our scheme and is discussed later), and krypton:

- \( \text{Ar} \rightarrow Z = 18 \), ground state \( \rightarrow 1s^22s^22p^63s^23p^6 \), ionization energy 15.759610 eV
- \( \text{Kr} \rightarrow Z = 36 \), ground state \( \rightarrow 1s^22s^22p^63s^23p^63d^{10}4s^24p^6 \), ionization energy 13.999605 eV
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Figure 2.2: Nonlinear process used to generate Lyman-α light. In this scheme the two-photon resonance-enhanced FWM (RE-FWM) uses the resonant condition of $\omega_1 = \omega_2$. For the generation of Lyman-α radiation at 121.56 nm, Kr gas is an efficient nonlinear medium since the 4s$^2$4p$^6 1S_0$ level is just $\sim 1300$ cm$^{-1}$ below the Lyman-α frequency, which adds another nearly resonant condition of $\omega_{jg} \sim 2\omega_1 - \omega_3$ in the third order susceptibility.
Figure 2.3: Grotrian diagram of Kr. The energy levels shown contribute to the nonlinear susceptibility for the RE-FWM process. Kr electron structure is described by \( j \ell \) coupling scheme an the term symbol is: \( ^{2S+1}I^P \) where \( P \) is the parity the term and \( \vec{K} = \vec{l} + \vec{j}_p \). Here \( \vec{j}_p \) is the angular momentum of the parent multiplet and \( \vec{l} \) that of the excited electron. The number next to each transition is the relative transition strength which have been compiled from Ref. [99, 100].
Any of the outermost electrons can transition to excited states. In all relevant excited states the structure remains of the form \((np^{5}n')\). A \((p^{5})\) structure is equivalent to \(p\) and it reduces to \((npn')\). This is equivalent to a two electron problem, one in \(p\) and the other on a \(s,p\) or \(d\) subshell. This behaviour is described by a \(Jj\) coupling where \(J\) is the total angular momentum quantum number for the parent ion and \(j\) is for the transitioning electron. A state in \(Jj\) coupling notation is written as \(|n_{j_{1}},j_{2}JM\rangle\), where \(n\) is the principal quantum number of the transitioning electron, \(j_{1}\) is the total angular momentum quantum number for the parent ion and \(j_{2}\) for the transitioning electron, \(J\) is the total angular momentum for the whole configuration, and \(M\) the magnetic quantum number referring to \(J\). The dipole transition selection rules are:

- \(\Delta j_{1} = 0, \Delta j_{2} = 0, \pm 1\) (but \(j = 0 \rightarrow j = 0\) is forbidden)
- \(\Delta J = 0, \pm 1\) (but \(J = 0 \rightarrow J = 0\) is forbidden)
- If \(M = 0\) and \(\Delta J = 0\) the matrix element is zero

In the \(jl\) scheme the electrostatic perturbation interaction produced by the the excited electron can be approximated as \(\vec{l}_{e}\cdot \vec{j}_{p}\). Here \(\vec{j}_{p}\) is the angular momentum of the parent multiplet and \(\vec{l}_{e}\) that of the excited electron. This electrostatic perturbation is diagonal in the Hilbert space \(|j_{p}l,K\rangle\), with \(K = \vec{l}_{e} + \vec{j}_{p}\). The coupling of the spin of the external electrons with parent ion is also diagonal as \(\vec{s}_{e} + \vec{j}_{p} = \vec{l}_{e} + \vec{s}_{e} + \vec{j}_{p} = \vec{K} + \vec{s}_{e} = \vec{j}\) where \(\vec{s}_{e}\) is the spin angular momentum of the excited electron. The energy split by spin-orbit coupling is then characterized by a couple of values \(j = K \pm 1/2\) and diagonal in the vector space \(|j_{p}l,K\rangle\). Any state can be uniquely determined by \(|nj_{p}l,Ks_{e}j\rangle\).

The states in Fig. 2.3 are represented using the term symbols for the \(jl\) coupling scheme, which have been used in this work for the calculations. The term symbol is: \(^{2S+1}[K]^{P}\) where \(P\) is the parity. For linear polarization only the \(z\) direction of the the dipole matrix elements \(r_{ab}\) is needed which can be obtained from tables of measured or tabulated values of oscillator strengths \(f_{ab}\), line strength, \(S(ab)\) or transition probabilities \(A_{ab}\) using the
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relations [101]:

$$|z_{ab}|^2 = |(a|z|b)|^2 = S(ab) = S(ba)$$  \hspace{1cm} (2.46)

$$A_{ab} = \frac{2\pi e^2}{m_e e_o A^2 g_b} f_{ab}$$  \hspace{1cm} (2.47)

$$f_{ab} = \frac{2}{3} \frac{\Delta E}{g_a} S(ab)$$  \hspace{1cm} (2.48)

where \(g_a\) and \(g_b\) are the statistical weights given by the degeneracy of the state \(g = 2J + 1\).

Using Eq. (2.28), and summing over all possible combinations of allowed transitions under \(j\ell\) coupling the susceptibility for the process has been calculated (see Fig. 2.4). The matrix elements were extracted from data tables [100, 102, 103] or tabulated from any available information using the previous relations. Notice that the uncertainty of oscillator strength measurements is between 20\(−\)50\% and this is reflected in the calculated susceptibility.

The two-photon resonance shape can be described by [104]:

$$S(2\omega_1) = \frac{2 \sqrt{\ln 2}}{\Delta \omega_{\text{Doppler}}} \sqrt{\pi} \int_{-\infty}^{\infty} \frac{\exp\left(-x^2\right)}{x - \zeta} \, dx,$$  \hspace{1cm} (2.49)

which is a scaled Plasma Dispersion function and where \(\zeta\) is:

$$\zeta = 2 \sqrt{\ln 2 \left(2\omega_1 + \frac{\Gamma_R}{2} - \omega_R\right) / \Delta \omega_{\text{Doppler}}}.$$  \hspace{1cm} (2.50)

Here \(\Gamma_R\) is the homogeneous Lorentzian linewidth of the resonant state \((2\pi \times 6.4 \text{ MHz})\). The plasma-dispersion function is equivalent to the Faddeeva function whose solutions are Voigt profiles (convolutions of a Lorentzian and a Gaussian profile).

Since Kr has several isotopes the two-photon resonance is actually composed of several resonance peaks. Details of the Kr isotopes can be seen in Table 2.2. Fig. 2.6 shows the calculated two photon resonance taking into
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Figure 2.4: Calculated third order susceptibility for RE-FWM process. Using Eq. (2.28), and summing over all possible combination of allowed transitions under $j l$ coupling, the susceptibility for the process have been calculated (see Fig. 2.4). The matrix elements were extracted from data tables [100, 102, 103] or tabulated from any available information using the previous relations. Notice that the uncertainty of oscillator strength measurements is between 20 – 50% and this is reflected in the calculated susceptibility.
Figure 2.5: Same as Fig. 2.4 where the calculated third order susceptibility for sum-difference and sum-frequency are shown in detail.
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account all the isotopes and the relative abundance. A comparison with a single isotope two-photon resonance indicates that the resonance strength is only reduced by $\sim 32\%$. The maximum difference between the resonance peaks of the individual isotopes is $0.0895 \text{ cm}^{-1} \sim 430 \text{ MHz}$. The linewidth of the resonance is one of the contributors to the theoretical intrinsic limit on linewidth for the generated wave in this process. There is also collisional broadening but the main practical limitation on the attainable linewidth are the fundamental beams. For pulsed Nd:YAG lasers, for instance, the linewidth is on the order of $\sim 5 \text{ GHz}$. There are, of course, various laser options with narrow linewidth but they usually require amplifier stages to reach the threshold power requirement for the nonlinear process.

Table 2.2: Kr stable isotopes with abundances of an earth bound mixture. The transition energy for the two-photon resonance, $5p^2 [1/2]$ state, is shown along with the transition energy for the two lowest electric dipole allowed transitions both around the Lyman-α energy.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Abundance</th>
<th>$5p^2 [1/2]$</th>
<th>$5s^2 [3/2]$</th>
<th>$5s^2 [1/2]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{78}\text{Kr}$</td>
<td>0.00355(3)</td>
<td>+ 0.0416</td>
<td>+ 0.6876</td>
<td>+ 0.7528</td>
</tr>
<tr>
<td>$^{80}\text{Kr}$</td>
<td>0.02286(10)</td>
<td>+ 0.0471</td>
<td>+ 0.6892</td>
<td>+ 0.7535</td>
</tr>
<tr>
<td>$^{82}\text{Kr}$</td>
<td>0.11593(31)</td>
<td>+ 0.0528</td>
<td>+ 0.6910</td>
<td>+ 0.7549</td>
</tr>
<tr>
<td>$^{83}\text{Kr}$</td>
<td>0.11300(19)</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>$^{84}\text{Kr}$</td>
<td>0.56987(15)</td>
<td>+ 0.0703</td>
<td>+ 0.7051</td>
<td>+ 0.7685</td>
</tr>
<tr>
<td>$^{86}\text{Kr}$</td>
<td>0.17279(41)</td>
<td>+ 0.1311</td>
<td>+ 0.7624</td>
<td>+ 0.8254</td>
</tr>
</tbody>
</table>

As the system is perturbed by the electromagnetic fields photons are absorbed and emitted. Some of the atoms will see two photons $\omega_R$ within $\tau_v \approx 0.68 \text{ fsec}$, the lifetime of the virtual state, and will excite to the $4p^5 (^{2}p_{1/2}) 5p^2 [1/2], J = 0$ level. When this happens the populations of the eigenstates will change. If a $\omega_T$ photon interacts with the atom within
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Figure 2.6: Calculated two-photon resonance ($5p^2[1/2]$) shape vs. detuning from the energy centre-of-gravity of the transition (weighted average of single isotope transition frequencies). The resonance of an ideal single isotope $^{84}$Kr gas mixture is shown for contrast. The resonance strength is reduced by ~32% and broadened due to the presence of the isotopes. Also the individual isotope transition frequencies and the abundance relative to $^{84}$Kr are shown.
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\( \tau_{5p} = 20.8 \text{ nsec} \) (the life time of the state) then a photon at \( \omega_i = \omega_{\text{Ly} - \alpha} \) the Lyman-\( \alpha \) line will be emitted. Otherwise, the photon will decay to another state generating parasitic electromagnetic fields corresponding to other processes.

The availability of atoms in the two photon resonant state limits the effectiveness of \( \omega_T \) photons bringing to completion the sum-difference mixing process. This gives an upper limit on \( I_3^o \) which, assuming the system is not near saturation, is a function of \( I_1^o \). Notice that having more \( \omega_T \) photons than this limit will not increase the \( \omega_{\text{Ly} - \alpha} \) photons generated. On the contrary, the excess of photons will only increase the probability for competing processes to take effect such as two-photon absorption and multi-photon ionization. This is discussed in the next section. The total power generated can be expressed as:

\[
\mathcal{P}_{\text{Ly} - \alpha} \propto N^2 \cdot \left[ \chi^{(3)} \right]^2 \cdot \frac{\gamma^2}{4} \cdot \mathcal{P}_R^3 \cdot F, \tag{2.51}
\]

where \( \gamma \) is a constant given by the requirement \( \lceil I_3^o \rceil = (1/\gamma)I_1^o \). The ceiling function of \( I_3^o \) indicates that providing more power than this maximum will not increase the power of the generated wave and the conversion efficiency will decrease. Based on the transition probabilities for transitions from the 5s states to the two-photon resonant state, then \( \gamma = 6.796 \). This is a very simplistic approximation but gives an indication of upper limit on the intensity \( I_3^o \). Providing any larger intensity will be counterproductive.

In rarified gases the density of the system \( N \) and the phase matching factor \( F \) will ultimately be responsible for the maximum efficiency achieved experimentally \( \eta = \mathcal{P}_{\text{Ly} - \alpha} / (\mathcal{P}_R + \mathcal{P}_T) \). However, all of the factors are intricately related. Phase matching is perhaps the most important aspect for nonlinear laser implementation. The phase matching coefficient is a measure of how efficiently the power is transferred from the input beams into the generated beam as shown. The maximum density possible is limited by parasitic processes and by phase matching. Furthermore, through phase matching it is possible to set up the environment so that competing processes, with different susceptibilities, are not favourable. Before discussing
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phase matching in more detail a brief analysis of the competing processes is given.

2.2.2 Competing and parasitic processes

As mentioned in the previous section some atoms will be excited to the two-photon resonant state. At this point they can interact with a $\omega_T$ photon or decay into the $4p^5(2p_{1/2})5s^2[1/2]$ or $4p^5(2p_{3/2})5s^2[3/2]$ states. All of these processes compete and the total perturbation can be expressed as an infinite perturbation expansion that included not only third order processes but also all the odd order processes allowed in a centrosymmetric medium. Furthermore, the description of power generation efficiency does not account for saturation effects at high powers. Factors contributing to saturation in the process include the Kerr effect, power broadening, stark shifts, multiphoton ionization, amplified spontaneous emission (ASE), parametric processes, hyper-Raman scattering, two photon absorption, self-defocusing, and higher (5th and 7th) order nonlinear effects. Fig. 2.7 shows some of the competing processes.

ASE involves emission from the two-photon resonant level $4p^5(2p_{1/2})5p^2[1/2], J = 0$ to the $4p^5(2p_{1/2})5s^2[1/2], J = 1$ level generating light at 557.45 nm or, with much lower probability, to the $4p^5(2p_{3/2})5s^2[3/2], J = 1$ level generating light at 768.74 nm. This process directly reduces the yield of Lyman-$\alpha$ photons. ASE presents some additional problems besides depleting the two-photon resonant level. The amplified light can power broaden the ASE transition modifying the $4p^5(2p_{1/2})5p^2$ state and further reduce the two-photon resonance. In addition, decay from the $4p^5(2p_{1/2})5s^2[1/2], J = 1$ and $4p^5(2p_{3/2})5s^2[3/2], J = 1$ states to ground will produce VUV light at 116.48 nm and 123.58 nm. This can be stimulated by the ASE completing a FWM process or could occur as a parametric process where the two photons (i.e. $\omega_{\text{idler}} = 557.45$ nm and $\omega_{\text{signal}} = 123.58$ nm) are generated. This presents a problem for detection as a bare solar blind photomultiplier would not be able to discriminate between Lyman-$\alpha$ and these VUV frequencies. ASE can be partially suppressed by phase matching as
Figure 2.7: Parasitic nonlinear processes that compete with the RE-FWM sum-difference process and reduces the yield of Lyman-α radiation.
the condition to generate Lyman-α radiation is different from the condition for 116.48 nm. However, 123.58 nm is very close and phase matching alone will not suppress it. Furthermore, the 123.58 nm transition is very strong with a transition probability of $3.12 \times 10^8 \text{ sec}^{-1}$ [100]. The only way to limit it is by providing enough $\omega_T$ photons. There is, however, a fine balance between providing sufficient $\omega_T$ photons to suppress ASE but below a threshold where the excess photons enhance two-photon absorption of the combination $\omega_{\text{Ly-α}} + \omega_T$.

Two-photon absorption (TPA) of the combination $\omega_{\text{Ly-α}} + \omega_T$ into the $4p^5(3p_{1/2})5p^{2}[1/2]$ state depletes the generated VUV radiation and leads to further parasitic processes. These type of multiphoton absorption mechanisms have been shown to be an important saturation mechanism [75, 76]. This absorption can be limited by controlling the intensity $I_T^0$ and reducing the interaction length (confocal length) of the tuneable gaussian beam $b_T$. The confocal length of $\omega_T$ should be shorter or equal than the one of the resonant beam $b_R$. This condition is easily achieved with TEM$_{00}$ beams.

Multiphoton ionization is a serious problem for high intensity or tightly focused beams. The ionization threshold for Kr is $\sim 14 \text{ eV}$ so multiphoton combinations such as $2\omega_R + \omega_T$ will lead to ionization. In addition, free electrons accelerated by the laser fields will collide with Kr atoms and lead to further pathways of ionization (see Fig. 2.7). The contribution of the ionization continuum to the refractive index may change the refractive index to the point that no conversion can occur. With pulsed lasers this is a problem that can be avoided by maintaining a loosely focused system (low intensity). At this point it is important to clarify some of the lexicon used in the literature when referring to tightly and loosely focused laser. In terms of the phase matching factor the tightly focused limit refers to the limit where $b/L \ll 1$ and this definition is only concerned with the dimensions of the terms on the phase matching integral. In terms of an optical process or photon-atom interaction it refers to some intensity threshold. This threshold is different for each system and is related to the availability of photons within time slots given by the uncertainty principle and the lifetimes of eigenstates of the system. It is possible to maintain low intensity
2.2. Source design and optimization

(or a loosely focused system) were the effects of saturation are negligible within the tightly focused limit of phase matching if the absorption is negligible (L can be made very long). In our case the threshold multiphoton ionization intensity limit for $I_0^R$ is on the order of $10^{12}$ W/m$^2$ based on an ionization cross section of 0.2 Mb for 6 eV photons in Kr [105] and we will operate below the limit where saturation effects become noticeable.

Raman processes stimulated by the generated Lyman-α are suppressed as there are no available even parity states for the stokes transition. Hyper-Raman processes involves a two-photon transition $\omega_R + \omega_{Ly\alpha}$ followed by scattering into any of the $d$ states. This process will increase the absorption of the generated Lyman-α light. However, multiphoton ionization will most likely occur before hyper-Raman scattering.

Finally, other saturation effects such as ac-stark shift and higher order processes will put an upper limit on the density and intensity of optical fields. This will result on a practical limit to the efficiency of conversion. In summary, phase matching is the principal tool to suppress parasitic processes and it is undesirable to work in the high intensity regime.

2.2.3 Phase matching

Phase matching will improve the conversion efficiency and will reduce parasitic effects by creating unfavourable conditions where energy and momentum are not conserved. In the RE-FWM process under discussion the optical depth (absorption) of the $\tau_R$, $\tau_T$ and $\tau_{Ly-\alpha}$ are negligible. Assuming plane waves and for a phase matched system with $\Delta k = 0$ Eq. (2.40) give us the phase matching factor. This expression reduces to:

$$F(\Delta kL = 0, \tau_R, \tau_T \ll 1, \tau_{Ly-\alpha} \ll 1) \approx 1 - \frac{\tau_R + \tau_T + \tau_{Ly-\alpha}}{2}. \quad (2.52)$$

For Gaussian beams, without considering optical absorption, Eq. (2.42) gives us the phase matching factor by performing the integral. For sum-difference Bjorklund [84] has already worked out the optimum condition which is $\Delta k b = 0$ (see Eq. (2.43)). However, this is only possible if $\Delta k = 0$, 

...
2.2. Source design and optimization

which is impossible to accomplish in a system with optical dispersion. Fig. 2.8 is the calculated wave vector mismatch for our process in Kr which is:

\[ CN = 2\pi \frac{(n_R - n_T - n_{Ly-\alpha})}{\lambda_{Ly-\alpha}}, \quad (2.53) \]

where \( C \) is the vector mismatch per atom and \( N \) is the number density. The index of refraction can be calculated from the linear standard Sellmeier formula:

\[ (n-1) = \frac{N r_e}{2\pi} \sum_i f_i \frac{\lambda_i^2}{\lambda_i^2 - \lambda^2}, \quad (2.54) \]

and the sum is performed over all the allowed transitions \( i \), \( r_e \) is the classical electron radius and \( f_i \) are the oscillator strengths. Fig. 2.9 shows the dispersion near the resonances of Kr and Ar and the index of refraction for \( \lambda_R \) and \( \lambda_T \). Also it shows the negative dispersion around the Lyman-\( \alpha \) region.

Fortunately, the condition \( \Delta k = 0 \) can be accomplished in a two-component system by “tuning” the effective index of refraction of the system. Two component phase matching in gases was first reported by Harris et al [71, 106, 107]. Effective index of refraction tuning by mixing two gases can accomplish the phase matching condition if the gases have opposite sign optical dispersion or if the generated wave is at least near a region with negative optical dispersion. In this case Kr has negative dispersion (or at least the dispersion turns negative) around the Lyman-\( \alpha \) region. Optimization is usually done by adjusting the ratio of the concentration of gases. If one of the components has no resonances near the optical fields applied then it only contributes to the linear dispersion and nonlinear effects on this medium do not play any role. The ratio of the two components can be
2.2. Source design and optimization

Figure 2.8: Calculated wavevector mismatch for the RE-FWM process in Kr where \( C \) is the vector mismatch per atom and \( N \) is the number density. This mismatch leads to destructive interference of the input beams and the generated Lyman-\( \alpha \) radiation.

Calculated using:

\[
\frac{N_a}{N_b} = \frac{\omega_{Ly-\alpha} \chi_b^{(1)}(\omega_{Ly-\alpha}) - \left[ 2\omega_R \chi_b^{(1)}(\omega_R) + \omega_T \chi_b^{(1)}(\omega_T) \right]}{2\omega_R \chi_a^{(1)}(\omega_R) + \omega_T \chi_a^{(1)}(\omega_T) - \omega_{Ly-\alpha} \chi_a^{(1)}(\omega_{Ly-\alpha})}. \tag{2.55}
\]

For a second component Ar gas has a suitable index of refraction and energy levels. Fig. 2.10 shows the Grotrian diagram of Ar and its ionization threshold [100]. In addition some of the parasitic processes that could occur in it are indicated.

As mentioned previously the oscillator strengths used for the susceptibility calculation have very large uncertainties and the calculated refractive index is not very accurate. It is possible to obtain a very accurate index
2.2. Source design and optimization

Figure 2.9: Calculated dispersion near the electric dipole resonances of Kr and Ar and the calculated index of refraction for $\lambda_R$ and $\lambda_T$. The negative dispersion around the Lyman-\(\alpha\) region is important for phase matching.
Figure 2.10: Argon's Grotrian diagram and some of the possible competing nonlinear processes. The lack of energy states in the energy region used for the nonlinear process makes it an ideal gas for phase matching mixing with Kr. The energy levels are based on Ref. [100].
of refraction for the gases using empirical three-point Sellmeier relations \cite{108} and this has been used to calculate \( n_R \) and \( n_T \). Unfortunately, the empirical Sellmeier relation does not predict the index of refraction in the region between the Kr resonances very well. Tabulated values for \( n_{Ly-\alpha} \) based on extrapolations of the linear Sellmeier relation adjusted to match experimental values \cite{108, 109} have been used to calculate \( n_{Ly-\alpha} \) and the optimal gas ratio. The tabulated values give two regions of gas mixtures where it is possible to accomplish phase matching. The mixture ratios Ar:Kr are between 3-4:1 and 16-18:1. Experimentally, it was found that the optimal ratio for the first region was Ar:Kr \( \approx 3.9:1 \) and for the second region was Ar:Kr \( \approx 17.7:1 \). However, the mixture ratio Ar:Kr \( \approx 3.9:1 \) produced ten times more power at \( \lambda_{Ly-\alpha} \) than at any other mixture. The index of refraction that are consistent with this first region of gas mixture ratio are shown in Table 2.3.

Table 2.3: Index of refraction from three-point Sellmeier relation of Kr and Ar used to calculate the phase matching mixture.

<table>
<thead>
<tr>
<th>Medium</th>
<th>( \lambda \Rightarrow 121.56 ) nm</th>
<th>202.31 nm</th>
<th>602.56 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kr</td>
<td>561.40</td>
<td>513.65</td>
<td>427.59</td>
</tr>
<tr>
<td>Ar</td>
<td>488.85</td>
<td>320.97</td>
<td>281.46</td>
</tr>
</tbody>
</table>

Now that the condition \( \Delta k = 0 \) can be accomplished, Eq. (2.43) indicates an optimal ratio \( b/L = 1 \). This is identical to the plane wave case. Without geometrical restrictions it is possible to use a loosely focused configuration. The loosely focused condition will assure that the intensity is below the saturation regime. In addition plane wave approximations are effective as the radius of the Gaussian wave fronts \( R_{Gaussian}(z < b) \rightarrow \infty \) and the interaction happens in a nearly plane wave region well within the Rayleigh
2.3. Third harmonic generation

length.

In summary, in this section a Lyman-α source based on FWM has been designed. The exact energy levels and two-photon resonance in krypton to be used for the process have been identified. The third order susceptibility for this process has been calculated and this indicates that the process should be efficient at generating VUV. Also the two-photon resonance shape has been calculated. An important result of this calculation is that even with the large number of stable krypton isotopes, the resonance broadening caused by the isotope shift does not cause a large reduction of the resonance strength (only ∼ 32%). A limit for the power of $\omega_T$ photons has been calculated indicating that the power of $\omega_T$ should be a maximum of ∼ 15% of the power of $\omega_R$ for efficient conversion. The most important parasitic process that can potentially occur, while reducing the conversion efficiency, have been identified. These processes are ASE, Parametric TPA and multiphoton ionization. Strategies to reduce their occurrence have been discussed. These strategies will be implemented and further discussed in the next chapter. Finally, the wavevector mismatch has been calculated and a strategy for phase matching has been identified. Phase matching requires the use of a gas mixture. The second gas in the mixture has been identified as argon and parasitic processes that could occur in this gas have been studied. The exact gas mixture ratio required for phase matching has also been calculated and is Ar:Kr ≈ 3.9:1. All of this information was used to implement the source and this is described in Chapter 3.

2.3 Third harmonic generation

Several Lyman-α sources have been designed based on THG and Table 2.4 is a compilation of sources using Kr as the nonlinear medium. Based on all the information available a non-resonant THG source was implemented in parallel with the RE-FWM implementation. Fig. 2.11 shows the energy diagram of the process.

Most of the optimal parameters in THG have been already worked out and the limits of conversion have been explored. The process has lower
## 2.3. Third harmonic generation

Table 2.4: Compilation of Lyman-\(\alpha\) sources implemented by tripling 364.8 nm and the reported efficiency of the conversion.

<table>
<thead>
<tr>
<th>Nonlinear medium</th>
<th>Phase matching</th>
<th>Efficiency (\eta)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kr</td>
<td>Ar</td>
<td>(10^{-5})</td>
<td>[58]</td>
</tr>
<tr>
<td>Kr</td>
<td>Ar</td>
<td>(10^{-4})</td>
<td>[59–62]</td>
</tr>
<tr>
<td>Kr</td>
<td></td>
<td>(10^{-5})</td>
<td>[63]</td>
</tr>
</tbody>
</table>

conversion efficiency than RE-FMW and the phase matching parameter space is much more restricted. However, THG is very attractive because it only requires one input color. This simplifies the implementation for the following reasons:

- Only one input beam is needed and no dichroic mirror is required to combine beams which is usually the most difficult optical item to obtain.
- The wavelength of the input beam is not in the UV region of the spectrum which results in less optical losses through the optical elements.
- In Kr there are less paths for parasitic processes and for instance ASE and multiphoton ionization is not a problem.
- A problem in FWM processes is the aligning of the different beams in the nonlinear medium due to dispersion and chromatic aberration of optical elements; this is not an issue in THG.
- Since only one beam is used, the temporal alignment of pulsed laser beams is not an issue.
- The phase matching condition could be easier to achieve as only two wavelengths are involved.

In addition there are some additional features that make this implementation attractive to the antihydrogen experiments with ALPHA:
2.3. Third harmonic generation

Figure 2.11: Energy diagram of non-resonant THG process used for the generation of Lyman-α radiation in Kr. The process uses only one input laser at 364.68 nm.

- The system requires less maintenance than a FWM based system as only one input laser is required.
- THG is a robust optical system as it does not require two collinear beams that would misaligned over time due to vibration.
- The system can be implemented with solid state lasers instead of dye lasers which require more maintenance.

THG does require high intensity for the three photon process to occur. This is usually accomplished by focusing the Gaussian beam. In the limit of tight focusing, \( b/L \ll 1 \), the limits to the phase matching integral of Eq. (2.42) can be approximated to \( L/2 \rightarrow \pm \infty \). The phase matching factor then
2.3. Third harmonic generation

reduces to:

\[
F(\Delta kL, b/L \ll 1) = \begin{cases} 
0 & \Delta kb \geq 0 \\
\pi^2 \left( \frac{b}{l} \right)^2 (\Delta kb)^2 \exp \{\Delta kb\} & \Delta kb < 0,
\end{cases}
\] (2.56)

which indicates that the wavevector difference should be negative \( \Delta k < 0 \). In fact the function goes through a maximum for \( \Delta kb = -2 \) and the phase matching factor becomes:

\[
F(\Delta kb = -2, b/l \ll 1) = \left( \frac{\pi b}{L} \right)^2 \exp \{ -2 \}. 
\] (2.57)

A two component mixture can also be used to find the phase matching condition in THG. A similar calculation as before predicts a gas mixture ratio of Ar:Kr \( \Rightarrow \) of 2.8:1 for \( \Delta k = 0 \). However, THG requires \( \Delta k < 0 \) which can be reached only with a mixture ratio lower than Ar:Kr \( \Rightarrow \) 2.8:1 and by adjusting the focus of the beam. In this implementation the optimal ratio was found experimentally and details of this are in the next chapter where the laser power measurements are discussed.
Chapter 3

Source implementation and characterization

Chapter 2 addressed the design of a Lyman-α source based on a RE-FWM process. The resonance enhancement is used as means of increasing the conversion efficiency of the process. RE-FWM process are reported to have a conversion efficiency $\eta$ on the order of \((\geq 10^{-4})\) \([67, 68, 110]\). This is an order of magnitude larger than typical THG processes (see Table 2.4). The particular two-photon resonance that is used in this work is particularly strong. This resonance coupled with the quasi-resonant third photon transition produces a very large third order susceptibility. We can expect a very efficient process if the phase matching condition is met and the parasitic processes are reduced.

As previously discussed, ASE and parametric processes also occur during the frequency mixing process generating $\lambda_{\text{parametric}} = 123.5$ nm in addition to the Lyman-α radiation $\lambda_{L\alpha} = 121.56$ nm. In order to measure the yield at the Lyman-α line it is necessary to separate all the spectral components. There are commercially available “Lyman-α” filters but the transmission peak is at 123.0 nm. Narrow filters have a typical transmission FWHM of 15.0 nm and a peak transmission of 19%. Furthermore, transmission through a narrow filter at 200.00 nm is still on the order of 1%. This means that in our RE-FWM process, assuming a maximum conversion efficiency of \(\geq 10^{-4}\), even a 0.1% of the $\omega_R$ photons surviving through the nonlinear process will be enough to bias a power measurement of the filtered signal. Some of the reports on Lyman-α sources have quoted power values based on direct measurements with filters. These measurements are obviously case dependent, but in general have large uncertainties associ-
ated with the use of the filters. In this particular case, with the detectors available, filters cannot be used. Part of the problem is that solar blind detectors, which use a Cs-I photocathode, are not characterized on the UV region of $\omega_R$ photons as these photons are supposed to be invisible. However, $\omega_R$ photons are not invisible. The different order of magnitudes of $\omega_{\text{Ly-}}$ and $\omega_R$ photons is such, that the detector signal for $\omega_{\text{Ly-}}$ cannot be discerned. The detector is blinded by the amount of $\omega_R$ photons. This is the problem and the reason this measurement is interesting. In this chapter the implementation of the Lyman-\(\alpha\) source and the power measurement are discussed.

During the implementation of the source the FWM process was optimized by changing the phase matching parameters (gas mixture, confocal length and interaction length) and the intensity of the input beams (via the focusing geometry). As a metric the power of the Lyman-\(\alpha\) radiation and light generated by the parasitic processes were monitored. These measurements were performed in vacuum using a diffraction grating with a photomultiplier tube (PMT) and this is discussed in the first section along with the optics and vacuum system implementation. To characterize the source, hydrogen was used as the detector and frequency discriminator. Laser Induced Fluorescence (LIF) of hydrogen was performed as means of measuring the effective power at the Lyman-\(\alpha\) source. The hydrogen for these measurements was produced with a commercial thermal gas cracker that is mostly used as a source for molecular beam epitaxy. The details of this experiment are discussed in the second section. Finally, as mentioned in the last section of the previous chapter, a THG source was developed in parallel to compare with the RE-FWM source. To measure the power of this source a detector based on the photoionization of acetone was developed and this is discussed in the last section.

### 3.1 Source implementation

Based on the results of the previous section a two-photon RE-FWM process was used to generate high power VUV coherent light. For this pro-
3.1. Source implementation

cess, the nonlinear medium is Kr gas. Here, two wavelengths ($\omega_R = 202.31$ and $\omega_T = 602.56$ nm) are mixed in a sum-difference scheme ($\omega_{\text{VUV}} = 2\omega_R - \omega_T$) with a two-photon resonance at ($4s^24p^55p[1/2]_o \leftarrow 4s^24p^6(1S_0)$), 98,855.1311 cm$^{-1}$ (101.1581 nm). At this wavelength it is possible to use LiF ($\sim$ 80% transmission) or MgF$_2$ ($\sim$ 50% transmission) windows and lenses. The most optimal optical layout is to have the two incoming TEM$_{00}$ mode beams collinear and with identical confocal parameters $b$. Figure 3.1 shows a schematic of the optical arrangement. A Nd:YAG laser (Spectra-Physics, Quanta-Ray Lab-130) producing light at the second harmonic, 532 nm (200 ± 60 mJ in a 8-12 ns pulse with 1 cm$^{-1}$ linewidth at 10 or 50 Hz), was used to pump two dye lasers to generate $\lambda_R \times 3 = 606.93$ nm (in a mixture of Rhodamine B and Rhodamine 101 dissolved in methanol) and $\lambda_T = 602.56$ nm (Rhodamine B in methanol) pulses. The 606.93 nm pulses were further frequency mixed in nonlinear crystals (BBO and KDP) to produce 0.2 ± 0.1 mJ of $\lambda_R = 202.31$ nm. The $\lambda_T = 602.56$ nm pulses were 0.05 ± 0.03 mJ. A dichroic mirror was used to combine the two pulses, and the combined beam was focused into the gas cell with a UV fused silica lens.
Figure 3.1: A Nd:YAG laser (Spectra-Physics, Quanta-Ray Lab-130) producing light at the second harmonic, 532 nm (200 ± 60 mJ in a 8-12 ns pulse with 1 cm⁻¹ linewidth at 10 or 50 Hz), was used to pump two dye lasers to generate $\lambda_R \times 3 = 606.93$ nm (in a mixture of Rhodamine B and Rhodamine 101 dissolved in methanol) and $\lambda_T = 602.56$ nm (Rhodamine B in methanol) pulses. The 606.93 nm pulses were further frequency mixed in nonlinear crystals (BBO and KDP) to produce $0.2 \pm 0.1$ mJ of $\lambda_R = 202.31$ nm. The $\lambda_T = 602.56$ nm pulses were $0.05 \pm 0.03$ mJ. A dichroic mirror was used to combine the two pulses, and the combined beam was focused into the gas cell with a UV fused silica lens.
The pump laser produced a beam diameter of 10 mm with a beam divergence of $< 0.5$ mrad. The dye laser generating the $\lambda_R \times 3 = 606.93$ nm (Narrowscan from Radiant Dyes) with a minimum linewidth of 0.04 cm$^{-1}$ $\sim 1200$ MHz and a wavelength tuneability of $\sim 0.02$ nm with wavelength stability is 0.001 nm/$^\circ$C. The output was fitted with a double Pellin Broka separation box to extract the ASE components generated in the dye. A telescope is used to compensate for differences in beam divergences between the beams. Collinearity is achieved by the use of the various mirrors in the path of $\omega_T$ (see Fig. 3.1). Temporal alignment of the pulses is achieved by adjusting the optical path of $\omega_R$ (by moving the laser and adjusting the propagation length) until the pulses of the two beams coincide. The $\lambda_T$ beam was generated in a Lambda Physik Scanmate OPPO dye laser which has a minimum linewidth of 0.1 cm$^{-1}$ $\sim 3$ GHz and a power stability of 10%. The output beams of both lasers were combined with a dichroic mirror. Two additional mirrors were used to align the two collinear beams, and the generated VUV beam, onto the detection system.

The nonlinear medium, a mixture of Kr:Ar was injected into a vacuum tube via flow meters. The tube was pre-pumped down to $10^{-6}$ Torr with a turbo pump. The pressure of the mixture was monitored using a capacitive pressure gauge. Both sides of the tube were sealed with o-ring fitted lens terminations suitable for plano-convex lenses. The incoming side received the UV fused silica lens and the output was fitted with a LiF window or a suitable MgF$_2$ lens according to the different measurements performed. The length of the nonlinear medium and the focal length of the input lens were optimized for the RE-FWM process.

As a first step the $\omega_R$ laser was calibrated to the two-photon resonant transition using a photoionization measurement of Kr. In resonant multiphoton ionization the atom absorbs photons and is excited to a quasi-stationary state (atom plus optical field) of the system. These resonances increase the ionization probability which is reflected on the photo-electron signal \[111\]. As a first order approximation the photoionization probability peak should coincide with the two-photon resonant transition. The $\omega_R$ laser was focused between two parallel plates 0.5 cm apart with a UV fused
silica lens of 10 cm focal length and in a chamber containing low pressure Kr gas (10 Torr). A voltage was applied between the plates between 100 V and 300 V. As the laser ionized Kr the field between the plates accelerated ions and electrons toward the plates and this resulted in a current measurement. The laser was tuned by maximizing the current measurement. The laser’s bandwidth estimated from this measurement is 0.1 cm\(^{-1}\) \(\sim\) 3 GHz. The \(\omega_T\) laser was initially calibrated using a laser wavelength meter (Coherent WaveMaster) with an accuracy of 0.005 nm \(\sim\) 4 GHz. Further calibration was performed by optimizing the fluorescence signal during LIF spectroscopy of hydrogen. As shown in the previous chapter (Fig. 2.6), the two-photon resonance is broad and has a complex shape due to the isotope shift. What this means is that all the laser \(\omega_R\) (with \(\sim\) 3 GHz linewidth) spectral energy takes part in the resonance transition. In terms of the expected linewidth of the generated Lyman-\(\alpha\) radiation, it can be estimated as the convolution of the \(\omega_R\) and \(\omega_T\) which are both Gaussian. The standard deviation of the convolution of two Gaussian functions \(f\) and \(g\) is:

\[
\sigma_{f \otimes g} = \sqrt{\sigma_f^2 + \sigma_g^2}.
\] (3.1)

The FWHM \(\Delta_{1/2}\) (linewidth) is related to \(\sigma\):

\[
\Delta_{1/2} = 2 \sqrt{2 \ln 2} \sigma \approx 2.35482 \sigma.
\] (3.2)

With an estimated linewidth for \(\omega_T\) of 3 GHz, the estimated linewidth of the Lyman-\(\alpha\) source is \(\sim\) 4.2 GHz. This is only an approximation as the full complexity of the two-photon resonance shape has not been considered.

A vacuum system was implemented to characterize the source. Fig. 3.2 show the schematic of the system which consists of a main vacuum chamber with a hydrogen source and a detector (solar blind photomultiplier tube Hamamatsu R972). The Lyman-\(\alpha\) chamber is attached to the laser input port and a home-built monochromator is attached to the laser output port. The best vacuum achieved was \(\sim\) 10\(^{-8}\) Torr with a 300 L/sec turbomolecular pump. The monochromator chamber has an additional
3.1. Source implementation

lens, diffraction gratings and pinhole. The same photomultiplier tube was swapped between ports for LIF experiments or source optimization with the monochromator.

Figure 3.2: Schematic of the vacuum system used to characterize the source. It consists of a main vacuum chamber with a hydrogen source and a detector (solar blind photomultiplier tube). The Lyman-α chamber is attached to the laser input port and a home-built monochromator is attached to the laser output port. The nonlinear medium, a mixture of Kr:Ar was injected Lyman-α chamber via flow meters. The pressure of the mixture was monitored using a capacitive pressure gauge. Both sides of the Lyman-α chamber were fitted with o-ring fitted lens terminations that are suitable for plano-convex lenses.

To separate the $\lambda_{param} = 123.5$ nm component from the Lyman-α radiation a simple monochromator was implemented. A plane ruled diffraction grating with 600 grooves/mm $\times$ 26 mm and a maximum resolving power
3.1. Source implementation

$\lambda/\Delta\lambda$ of 15600 was used. The grating equation:

$$m\lambda = d(\sin \alpha \pm \sin \beta), \quad (3.3)$$

is used to calculate the angles $\alpha$ (incident) and $\beta$ (diffracted). In the equation $d$ is the groove spacing given by the manufacturer and $m$ is the diffraction order which in this case is negative. Fig. 3.3 shows a picture of the setup. The angle of the grating was manipulated using a home-built tilt manipulator which was o-ring sealed. With the o-ring sealed manipulator it was possible to reach a vacuum of $\sim 10^{-6}$ Torr. With collimating optics at the output of Kr cell and a pinhole 30 cm away from the grating and in front of the photomultiplier tube it was possible to discriminate between $\lambda_{\text{param}} = 123.5$ and $\lambda_{\text{Ly} - \alpha} = 121.56$. In addition, the interior of the vacuum tube was covered with blackened foil to reduce reflections.

This experimental setup was not suitable for quantitative measurements, but it was used to optimize the phase matching geometry (L, b and mixture ratio) qualitatively by comparing the power of $\lambda_{\text{param}} = 123.5$ and $\lambda_{\text{Ly} - \alpha} = 121.56$. Two different ratios of the gas mixtures were used and three different focusing geometries were compared. Table 3.1 shows the qualitative results.

The most optimal configuration was using a nonlinear medium length $L = 98.9$ cm, an input lens focal length of 500 mm and a gas mixture of Ar:Kr of 3:1 with 100 mTorr of Kr. The gas mixture was further optimized in this configuration to Ar:Kr 3.9:1 and 100 mTorr of Kr. The $\omega_L$ laser beam had a $1/e^2$ radius at the input laser of $\sim 0.75$ mm. It is possible to calculate the Gaussian beam parameters using the relations:

$$\frac{1}{q} = \frac{1}{R} - \frac{i\lambda}{\pi W^2} \quad (3.4)$$

and

$$q^2 = \frac{Aq_1 + B}{Cq_1 + D}, \quad (3.5)$$

where $R = z(1 + z_0^2/z^2)$ is the radius of curvature, $W = W_0 \sqrt{1 + z^2/z_0^2}$ is the
3.1. Source implementation

![Source implementation diagram]

Figure 3.3: Source characterization chamber with a simple home-built monochromator. A plane ruled diffraction grating with 600 grooves/mm × 26 mm and a maximum resolving power $\lambda/\Delta\lambda$ of 15600 was used.

Initial beam radius and $z_0 = \pi W_0^2 / \lambda$. For propagation through a lens $A = 1, B = 0, C = -1/f$ and $D = 1$; for propagation through air $A = 1, B = d$ (distance), $C = 0$ and $D = 1$. The beam radius at the focal point is calculated this way to be 40 µm and the confocal length $b$ is 49.67 mm. As mentioned, this calculation only applies to Gaussian beams and the beams generated by the dye laser have transverse modes that are not necessarily Gaussian. Since a beam profiler was not available, the transverse modes were not characterized. However, by visual inspection no multiple transverse modes were identified. The UV fused silica lens has a transmission of 93% for 602 nm.
3.1. Source implementation

Table 3.1: Optimization of phase matching parameters. The cell length L, input lens focal length f and gas mixture ratio where optimized by measuring the power of $\lambda_{Ly-\alpha} = 121.6$ nm and the parasitic $\lambda_{param} = 123.5$ nm. The wavelength discrimination was done with a diffraction grating in the setup shown in Fig. 3.3. An optimal configuration of $L = 28.9$ cm, $f = 500$ mm and gas mixture of Ar:Kr of 3.9:1 was adopted for the remaining experiments.

<table>
<thead>
<tr>
<th>L (cm)</th>
<th>f (mm)</th>
<th>Ar:Kr Ratio</th>
<th>$\lambda_{Ly-\alpha}$ = 121.6 nm</th>
<th>$\lambda_{param}$ = 123.5 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>36.5</td>
<td>200</td>
<td>3:1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>200</td>
<td>16:1</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>56.9</td>
<td>300</td>
<td>3:1</td>
<td>8</td>
<td>1</td>
</tr>
<tr>
<td>300</td>
<td>16:1</td>
<td>5</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>56.9</td>
<td>500</td>
<td>3:1</td>
<td>10</td>
<td>0.2</td>
</tr>
<tr>
<td>500</td>
<td>16:1</td>
<td>1</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

and < 90% for 202 nm. The average $\omega_R$ intensity at the focal point with a repetition rate of 10 Hz is $3.5 \times 10^5$ W/m². This corresponds to a peak intensity with a 10 nsec pulse of $3.5 \times 10^{13}$ W/m². This calculation does not consider the absorption of the laser before the focal point. The $\omega_T$ laser beam had a $1/e^2$ radius at the input laser of $\sim 2.0$ mm. The beam radius at the focal point is calculated to be $50 \mu$m and the confocal length b is 26.06 mm. Fig. 3.4 shows the configuration where the chromatic aberration of the input lens is not considered. Finally, similarly to the Kr ionization experiment, two bias plates were placed on the Lyman-$\alpha$ generation chamber to monitor ionization around the focal point. A very sporadic photoionization signal could be measured when the plates where biased to 100 V and mostly due to power fluctuations of the Nd:YAG pump laser. Otherwise, normal operation was below the photoionization threshold.
3.1. Source implementation

Figure 3.4: Schematic of the incoming beams’ intensity profiles in the Lyman α cell. The most optimal configuration was using a nonlinear medium length $L = 98.9$ cm, an input lens focal length of 500 mm and a gas mixture of Ar:Kr of 3.9:1.
3.2 Lyman-α source characterization

In the previous section the implementation and optimization of the source were discussed. The two-photon resonant laser was calibrated by maximizing the photoionization current generated by focusing the laser on low pressure Kr. The phase matching parameters were optimized using a home-built monochromator by comparing the power of the light generated by the RE-FWM (Lyman-α) and parasitic processes in different configurations. However, the home-built monochromator did not provide enough repeatability to be used for quantitative measurements. Furthermore, with the equipment implemented, it is not possible to do an absolute power measurement since there is not enough information to calculate the losses. Instead, to measure the Lyman-α power, LIF of hydrogen was performed. The Lyman-α photons excite hydrogen to the 2p state. Electrons then decay to the ground state and emit Lyman-α photons. However, the emission is omni-directional, and a detector at right angle from the laser would only be able to see some of the fluorescence. This is the basis of LIF power measurement. The hydrogen for these measurements was generated using a commercial source. In this section the LIF experiment and the absolute Lyman-α power calculated from those measurements are discussed.

3.2.1 Effusive hydrogen source spectroscopy

Atomic hydrogen was generated by thermal dissociation of H₂ with a Mantis MGC75 thermal gas cracker. The molecular hydrogen was flown through the thermal gas crackers heated capillary (tube of 2.5 mm inside diameter) at a maximum rate of 1.0 sccm to generate atomic hydrogen. Hot atomic hydrogen emerging from the tip of the capillary was irradiated with the Lyman-α source pulses. The fluorescence and absorption of Lyman-α light were measured with a solar blind photomultiplier.
3.2. Lyman-α source characterization

Hydrogen source

The gas cracker uses a biased, hot tungsten filament to generate electrons and accelerate them towards an iridium capillary. Electron bombardment heats up the capillary to 1500 K. Molecules flown through the capillary are dissociated by collisions with the wall. The whole arrangement is encased on a water cooled block. According to measurements provided by the manufacturer it is possible to achieve up to 90% dissociation of hydrogen with a gas flow of up to 1.0 sscm. A published independent characterization of the device reported a hydrogen flux of $1.3 \times 10^{17}$ atoms / (sec $\cdot$ cm$^2$) impinging on a sample of highly ordered pyrolytic graphite [112]. The measurement was done by characterizing the profile of chemical etching caused by hydrogen on the graphite. The chamber pressure was $10^{-4}$ Torr which, using the Hertz-Knudsen formula for incident flux $F$:

$$F = \frac{p}{\sqrt{2\pi mk_B T}},$$

is equivalent to an incident flux of $1.58 \times 10^{17}$ atoms / (sec $\cdot$ cm$^2$). This measurement indicates a steady state atomic density of $\sim 83\%$ hydrogen in the chamber.

Fig. [3.5] shows the experimental setup used. For LIF experiments a MgF$_2$ lens with a focal length of 200 mm focused the laser directly in front of the gas cracker at 2.5 cm from the capillary. The photomultiplier tube was at right angle and assuming a point source for the fluorescence, it captured a $2.2485 \times 10^{-4}$ of the sphere. The hydrogen in front of the source can be assumed to be thermalized at the temperature of the capillary and is, therefore, very hot at $\sim 1500$ K. The velocity distribution of the hydrogen in the chamber is given by the Maxwell-Boltzman distribution:

$$f(v) = \frac{4\pi}{2\pi k_B T} \left[ \frac{m}{2\pi k_B T} \right]^{3/2} v^2 \exp\left\{ \frac{-mv^2}{2k_B T} \right\}.$$  (3.7)

For hydrogen the mean speed $\bar{v} = \sqrt{(8k_B T)/(\pi m)}$ is 5635.52 m/s. The ra-
3.2. Lyman-α source characterization

diative lifetime of the $2p$ state of hydrogen is:

$$\tau_k = \sum_i A_{ki},$$  \hspace{1cm} (3.8)

where $A_{ki}$ are the transition probabilities to all levels $i$ lower in energy than $k$. For hydrogen $\tau_{2p} = 2.128 \text{ nsec}$ based on published transition probabilities [100]. In the chamber, an atom would have moved $12 \mu\text{m}$ between absorption of the photon and the fluorescence which is not a problem for the LIF experiment as the view area of the detector is larger. The actual spot size of the Lyman-α source is unknown. However, it is reasonable to assume that the laser has approximately the same Gaussian beam parameters as $\omega_R$ and most of the fluorescence is produced at the focal point of the lasers. Following this assumption the Lyman-α source would have a radius of $2 \text{ mm}$ at the focusing lens, a waist radius less than $10 \mu\text{m}$ and confocal length $b \approx 20 \mu\text{m}$. Given these dimensions, it is assumed that the fluorescence is point-like and the detector captures all of the photons within the solid angle viewing area ($2.2485 \times 10^{-4} \times 4\pi$). The chamber is filled with hydrogen and there is absorption (and fluorescence) of Lyman-α before the focal point of the laser, but this is not considered. Finally, the absolute power measurement based on fluorescence has a large uncertainty, due to the described assumptions. However, it is possible to measure a lower bound for the Lyman-α power, and this bound is really what is required to evaluate the source.
3.2. Lyman-α source characterization

Figure 3.5: For LIF experiments a MgF₂ lens with a focal length of 200 mm focused the laser directly in front of the gas cracker at 2.5 cm from the capillary. The photomultiplier tube was at right angle and assuming a point source for the fluorescence, it captured a $2.2485 \times 10^{-4}$ of the sphere. The hydrogen in front of the source can be assumed to be thermalized at the temperature of the capillary and is, therefore, very hot at $\sim 1500$ K.
3.2. Lyman-α source characterization

Hydrogen spectroscopy

To measure fluorescence, the light scattered was recorded with the PMT. A photodiode (activated by the $\omega_T$) was used to trigger the data collection. The PMT uses a MgF$_2$ window and a Cs-I photocathode. Fig. 3.6 shows the PMT pulses and the photodiode signal. Unfortunately, Cs-I is still activated by $\omega_R$, which is at the threshold of its spectral range (115 nm - 200 nm). Most of the PMT pulse is caused by scattered $\omega_R = 202.38$ nm photons. However, when the hydrogen source is turned on and off it is possible to see a small difference caused by Lyman-α fluorescence. During the experiments the molecular hydrogen was continuously flowing and only the bias between the tungsten filament and the capillary was switched on and off. The pulses of Fig. 3.6 are the result of 128 averages. The actual fluorescence signal was extracted by subtracting the pulses with the hydrogen source on and off. The total energy per pulse is proportional to the integrated fluorescence signal also shown at the bottom of Fig. 3.6. The total length of the PMT signal pulse is 23.8 nsec, which is much longer than the expected Lyman-α pulse length due to the detector response time.

PMT’s can be modelled as capacitive-resistive (CR) circuits followed by stages of RC circuits. CR stages are differentiators and RC are integrators. The time domain output signal for a delta pulse input signal to an RC-CR circuit is:

$$V_{out}(t) = \frac{V_0 \tau_1}{\tau_1 - \tau_2} \left( \exp\left(-\frac{t}{\tau_1}\right) - \exp\left(-\frac{t}{\tau_2}\right) \right),$$

where $\tau$ are the time constants and $V_0$ is the input voltage. Fig. 3.7 shows the time domain response for a single RC-CR circuit. For a large number of stages the output pulse becomes Gaussian. For the Hamamatsu PMT there are 11 stages. The rise time (time for the output signal to go from 10% to 90% given a delta pulse input) is 1.6 nsec. The electron transit time is 17 nsec and the maximum gain is $10^5$. For the linear-focused PMTs (the type used here) the expected pulse width (FWHM) of the response function is 5 nsec. The standard deviation $\sigma_{PMT}$ is 2.12 nsec. To calculate the input pulse shape, it must be de-convoluted from the output PMT.
3.2. Lyman-α source characterization

Figure 3.6: To measure fluorescence the light scattered was recorded with the PMT. A photodiode (activated by the $\omega_T$ pulse) was used to trigger the data collection. Most of the PMT pulse is caused by scattered $\omega_R = 202.38$ nm photons. However, when the hydrogen source is turned on and off it is possible to see a small difference caused by Lyman-α fluorescence. The pulses are the result of 128 averages. The actual fluorescence signal was extracted by subtracting the pulses with the hydrogen source on and off. The total energy per pulse is proportional to the integrated fluorescence signal shown at the bottom. The total length of the PMT signal pulse is 23.8 nsec, which is much longer than the expected Lyman-α pulse length due to the detector response time.
3.2. Lyman-α source characterization

signal using the calculated response function of the photomultiplier. The de-convolution of two Gaussians is another Gaussian and using Eq. (3.2) is possible to extract the standard deviation. The PMT signal can be fitted to a Gaussian with a standard deviation $\sigma_{\text{signal}} = 5.12$ nsec and the PMT $\sigma_{\text{PMT}} = 2.12$ nsec. The laser pulse (combined $\omega_R$ and Lyman-α photons) $\sigma_{\text{Laser}} = 5.0$ and the FWHM is 11.9 nsec. The Lyman-α (fluorescence) pulse is smaller with a FWHM of 9 nsec.

In addition to the power measurement, LIF was also used to perform a crude measurement of the Lyman-α linewidth. To calibrate the $\omega_T$ the fluorescence signal was measured with the PMT as the wavelength was swept. The peak of the signal correlates with the power of the generated Lyman-α radiation. These measurements also provided a line shape used to estimate the source linewidth. As mentioned before, the hydrogen in the chamber directly in front of the the source has a mean speed of 5635.5 m/sec and this produces Doppler broadening:

$$\Delta\omega = \frac{2\omega_0}{c} \sqrt{2 \ln \frac{2k_BT}{m}}.$$  

The Doppler broadening for hydrogen is 68.42 GHz at FWHM. The natural transition line shapes of atoms have a Lorentzian profile and for the hydrogen ($1s - 2p$) transition the linewidth of this profile is 99.7 MHz. Thermal broadening produces an absorption line with a Gaussian profile. The net profile is the convolution of both profiles, a Gaussian and a Lorentzian, which is called a Voigt profile. The Voigt profile is dominated by the Gaussian shape around the mean and by the Lorentzian in the wings (tails). In this case the thermal broadening linewidth is approximately three orders of magnitude larger than the natural linewidth and the net line shape can be assumed to be Gaussian with a FWHM of 68.43 GHz which is 2.281 cm$^{-1}$. The measurements shown in Fig. 3.8 were fitted with a Gaussian that is assumed to mimic the line shape of the transition. The measurement at the peak of the Gaussian in Fig. 3.8 corresponds to the fluorescence recorded in Fig. 3.6 with a peak fluorescence signal of 14.9 mV. The standard deviation of the measured transition shape is $\sigma_{\text{measured}} = 0.9725$ cm$^{-1}$ and
3.2. Lyman-α source characterization

Figure 3.7: PMT’s can be modelled as capacitive-resistive (CR) circuits followed by stages of RC circuits. CR stages are differentiators and RC are integrators. For a large number of stages the output pulse becomes Gaussian. To calculate the input pulse shape, it must be de-convoluted from the output PMT signal using the calculated response function of the photomultiplier. The de-convolution of two Gaussians is another Gaussian and using Eq. (3.2) is possible to extract the standard deviation. The PMT signal can be fitted to a Gaussian with a standard deviation $\sigma_{\text{signal}} = 5.12$ nsec and the PMT $\sigma_{\text{PMT}} = 2.12$ nsec. The laser pulse (combined $\omega_R$ and Lyman-α photons) $\sigma_{\text{Laser}} = 5.0$ and the FWHM is 11.9 nsec. The Lyman-α (fluorescence) pulse is smaller with a FWHM of 9 nsec.
3.2. Lyman-α source characterization

the standard deviation corresponding to a linewidth of 68.43 GHz is \( \sigma_H = 0.968 \text{ cm}^{-1} \). It is possible to de-convolute the source line shape from the measured spectrum using the calculated line shape of the hot hydrogen. The calculated linewidth is 6.6 GHz which is 2 GHz larger than the original estimated from the \( \omega_T \) input laser bandwidth of \( \sim 4 \text{ GHz} \). This is most likely due to the broadening of the two-photon resonance caused by collisions (pressure broadening), power broadening and ionization of Kr and the linewidth of the \( \omega_R \).

Finally, the maximum power measured for the Lyman-α source was calculated from the integrated fluorescence signal as shown in Fig. 3.6. Based on an anode minimum sensitivity at \( \Lambda_{Ly-\alpha} \) of 200 A/W the power of the Lyman-α radiation is:

\[
0.125 \pm 0.030 \mu J \text{ @ 10 Hz and 9 nsec pulses: } 1.25 \pm 0.30 \mu W.
\]

This power assumes that every Lyman-α photon is being scattered by hydrogen. The uncertainty of the PMT gain is not known and very little information about it has been published. It could potentially be quite large as the gain in region of operation is not linear. The peak power delivery per pulse 12.45 W and the area at the Lyman-α beam focus was estimated to be \( 3.14 \times 10^{-10} \text{ m}^2 \). The intensity is \( 39.63 \times 10^9 \text{ W/m}^2 \). The conversion efficiency of the process is \( \eta \sim 6 \times 10^{-4} \) which agrees with the reported efficiencies on the order of \( \geq 10^{-4} \) \([67, 68, 110]\). The power generated meets the requirements set for antihydrogen cooling. Power measurements were performed also at 50 Hz repetition rate as it was thought that the higher repetition would yield higher average power. However, at 50 Hz the Nd:YAG laser could not deliver as much energy per pulse. The maximum power measured for 50 Hz is:

\[
0.025 \pm 0.006 \mu J \text{ @ 50 Hz and 9 nsec pulses: } 1.25 \pm 0.30 \mu W.
\]

These power measurements represent the maximum obtained after care-
3.2. Lyman-α source characterization

Figure 3.8: The net line shape of the hydrogen $1s-2p$ transition can be assumed to be Gaussian with a FWHM of 68.43 GHz which is 2.281 cm$^{-1}$. Fluorescence measurements performed as $\omega_T$ was swept where fitted with a Gaussian that is assumed to mimic the line shape of the transition. The measurement at the peak of the Gaussian corresponds to the fluorescence recorded in Fig. 3.6 with a peak fluorescence signal of 14.9 mV. The standard deviation of the measured transition shape is $\sigma_{\text{measured}} = 0.9725$ cm$^{-1}$ and the standard deviation corresponding to a linewidth of 68.43 GHz is $\sigma_H = 0.968$ cm$^{-1}$. It is possible to de-convolute the the source line shape from the measured spectrum using the calculated line shape of the hot hydrogen. The calculated linewidth is 6.6 GHz which is 2 GHz larger than the original estimated from the $\omega_T$ input laser bandwidth of $\sim$ 4 GHz. This is most likely due to the broadening of the two-photon resonance caused by collisions (pressure broadening) and ionization of Kr and the linewidth of the $\omega_R$. 

$\nu_T$ (cm$^{-1}$) Relative to Calibration
ful optimization and capture the potential of the system. The system is not stable in the sense that it requires careful daily optimization before operation and changes hour-by-hour. Power fluctuations presumably due to temperature in the pump and dye lasers make these measurements challenging. Also the nonlinear crystals change during the measurement run, presumably due to temperature gradients produced by the laser, and this is reflected in the output UV light power. The density of hydrogen is high ($\sim 10^{-4}$ Torr) so it is assumed all the Lyman-$\alpha$ photons are absorbed, which is reasonable given the cross section for this radiation is very large. As mentioned previously, the high temperature of the hydrogen causes large thermal broadening of 62.48 GHz. The laser linewidth was calculated to be $\sim 6$ GHz which is about a tenth of the linewidth of the hydrogen and for that reason efficient at inducing fluorescence (all the photons can interact with some velocity subset of atoms). It is interesting to perform the same experiment with cold hydrogen. However, hydrogen at 1 K already has a Doppler broadening of 1 GHz. To reduce the Doppler broadening of the hydrogen, or to “cool” the hydrogen, there are well established molecular beam techniques. In the next chapter the implementation and characterization of a supersonic beam of hydrogen is described.

3.3 Implementation of THG Lyman-$\alpha$ source

As mentioned previously, the conversion efficiency of the RE-FWM process is higher than that of THG. However, implementation of THG is easier since only one laser is required. Even though the phase matching condition for THG is more restricted than other FWM processes, it is more forgiving with respect to parasitic processes. As mentioned before there are no multiphoton ionization, multiphoton absorption or ASE parasitic processes occurring during the Lyman-$\alpha$ generation. Power measurements are also simpler since the input laser is easily discriminated. For instance, two Lyman-$\alpha$ filters in series with a transmission of $< 0.1\%$ at 364.7 nm and a solar-blind photomultiplier would be sufficient for a power measurement of the Lyman-$\alpha$ light generated. In this section the implementation and
characterization of the THG source is described.

3.3. Implementation of THG Lyman-α source

3.3.1 Source implementation

The THG of Lyman-α requires an input laser $\lambda_{THG} = 364.68$ nm. To generate it an Nd:YAG laser (Spectra-Physics, Quanta-Ray Lab- 130) producing light at the second harmonic, 532 nm (200 ± 60 mJ in a 8-12 ns pulse with 1 cm$^{-1}$ linewidth at 10 Hz), was used to pump a dye laser (Lambda Physik Scanmate OPPO) using Rhodamine B in methanol. This generated pulses of 729.36 nm light which were further doubled in a BBO crystal producing $\lambda_{THG} = 364.68$ nm pulses of $\sim 2 \pm 0.5$ mJ. The laser was focused into a chamber with a Kr:Ar mixture using a fused silica lens with $f = 300$ mm. The cell length $L$ was 56.9 cm and the $1/e^2$ radius of the beam at the lens was 2 mm. The radius at the focus is calculated to be 20 $\mu$m. This is equivalent to a peak intensity of $1.5 \times 10^{15}$ W/m$^2$ which is two orders of magnitude higher than the peak intensity of the RE-FWM process. This is required to drive the three-photon process. The optimization of the gas mixture was done by measuring the power of the generated VUV light. Calibration of the 729.36 nm light was done with the wavelength meter (Coherent Wave-Master) with an accuracy of 0.005 nm. At these power levels for $\lambda_{THG}$ it was thought that the incoming photons could induce a three-photon resonant process on hydrogen generating fluorescence at Lyman-α. For this reason LIF was not used to measure the power of the THG source. Furthermore, the power of the $\lambda_{THG}$ laser was beyond the damage threshold of the diffraction grating which is only a couple of centimetres squared. For these reasons, to measure the absolute power an acetone ionization detector was implemented.

3.3.2 Acetone ionization detector

Acetone (CH$_3$)$_2$CO has been previously investigated using high resolution VUV synchrotron radiation in the energy ranges from 3.7 - 10.8 eV [113]. The photo-absorption spectrum is well know and there are reliable measurements of the cross section. Furthermore, high purity acetone of 99.8%
3.3. Implementation of THG Lyman-α source

is readily available. The ionization threshold of acetone has been investigated by mass-analyzed threshold ionization and zero-kinetic energy techniques [114]. The ionization energy is 78299.6 cm\(^{-1}\) and the absorption cross section for Lyman-α radiation is \(\sigma_{Ly-\alpha} = 38\) Mb. The THG input laser \(\lambda_{THG} = 364.68\) nm is equivalent to an energy of 3.3998 eV (27422 cm\(^{-1}\)). These photons are well below the ionization threshold and the photoabsorption cross section is \(\sim 0.01\) Mb. For these reasons acetone is very promising as a Lyman-α radiation power detector for the THG process.

To implement a detector a vacuum chamber fitted with a LiF window was connected to the Lyman-α cell. The acetone chamber was fitted with two parallel plates 20 cm long and 2 cm wide separated by 1 cm. The laser propagated along the cell between the plates. The plates were biased up to 500 V. A tube containing the liquid acetone sample is connected to the vacuum chamber and the acetone was degassed by repeated freeze-pump-thaw cycles. The acetone was frozen by submerging the tube in liquid nitrogen. After a few cycles the acetone was frozen once more and the chamber pumped down to \(\sim 10^{-6}\) Torr. After the acetone thaws it fills the chamber with acetone vapour and the pressure is monitored with a pressure gauge. Since acetone vapour pressure is \(\sim 100\) Torr at room temperature, the density inside the cell is high. When the laser propagates between the bias plates, it is possible to measure the current generated by the photo-electrons. The electron count is proportional to the Lyman-α power:

\[
\Gamma_e = \Phi \sigma_{Ly-\alpha} N,
\]  

(3.11)

where \(\Gamma_e\) is the electron count per time volume, \(\Phi\) is the photon flux and \(N\) is the molecule number density. Lyman-α power measurements were performed with the acetone detector as the gas mixture was varied. Fig. 3.10 shows the power measurement and the optimal gas mixture.
3.3. Implementation of THG Lyman-α source

Figure 3.9: Acetone (CH$_3$)$_2$CO was used to detect Lyman-α radiation. The ionization energy is 78299.6 cm$^{-1}$ and the absorption cross section for Lyman-α radiation is $\sigma_{Ly-\alpha} = 38$ Mb. $\lambda_{THG} = 364.68$ nm is well below the ionization threshold. The acetone chamber was fitted with two parallel plates 20 cm long and 2 cm wide separated by 1 cm. The laser propagated along the cell between the plates. The plates were biased up to 500 V. A tube containing the liquid acetone sample is connected to the vacuum chamber and the acetone was degassed by repeated freeze - pump - thaw cycles. When the laser propagates between the bias plates, it is possible to measure the current generated by the photo-electrons. The electron count is proportional to the Lyman-α power.
3.3. Implementation of THG Lyman-α source

Figure 3.10: Power measurements performed in an acetone detector. The laser propagated along the acetone cell between the biased plates. Photoelectrons generated by the Lyman-α photons and based on the acetone cross-section of \( \sigma_{\text{Ly-}\alpha} = 38 \text{ Mb} \) the power was calculated. The ionization energy of acetone is 78299.6 \( \text{cm}^{-1} \), well beyond the \( \lambda_{\text{THG}} \) energy of 27422 \( \text{cm}^{-1} \). During the measurement Kr was added first to the mixing cell and then Ar. The increase in power is due to better phase matching conditions.
3.3. Implementation of THG Lyman-α source

The maximum power generated (∼ 0.6 µW) indicates a conversion efficiency $\eta \sim 3 \times 10^{-5}$. Since the THG source was only implemented for comparison with the RE-FWM source, no more measurements were performed. An advantage of the THG process is that the input power can potentially be increased more than in the RE-FWM case. This is due to the fact that the threshold intensity for photoionization of Kr is much higher for the $\lambda_{THG}$ photons than for the two-photon resonant $\lambda_R$ photons. Also there is very little losses in the optics for the THG system. In Chapter 5 the two systems are compared and the current work on the THG source is discussed.
Chapter 4

Hydrogen beam implementation

In the previous chapters the design, optimization and implementation of a Lyman-α source were discussed in detail. Although the development of the source is the main focus of this dissertation, an equal or even larger effort has been put into the development of a cold hydrogen beam. In the introductory chapter it was mentioned that in order to cool hydrogen it was estimated that the source would require a linewidth on the order of \( \sim 100 \) MHz and a power of \( \sim 1.0 \) \( \mu \)W. The hot hydrogen LIF experiment described in Chapter 3 indicated that the source could potentially be used for the antihydrogen experiment as the power threshold was met. However, LIF on a cold hydrogen beam would confirm that the source could detect and, subject to linewidth reduction, even cool antihydrogen. It is important to point out that the main limitations on the bandwidth of the generated laser are the linewidth of the input lasers. The high temperature of the hydrogen dissociated by the gas cracker causes large thermal broadening of 62.48 GHz. The laser linewidth was calculated to be \( \sim 6 \) GHz which is about a tenth of the linewidth of the hydrogen transition and for that reason it is very efficient at inducing fluorescence. However, hydrogen at 1 K already has a Doppler broadening of 1 GHz. To reduce the Doppler broadening of the hydrogen transition, or to “cool” the hydrogen, we have developed a supersonic hydrogen beam. The implementation of the beam is based on the well established techniques of supersonically expanded molecular beams. In this chapter the implementation of the beam and its characterization are discussed. The first section is a very short summary of the theory of supersonic beams. This is followed by a description of the hardware
4.1 Supersonic beams

A beam of cold atoms can be obtained through a supersonic jet expansion mechanism. The gas expansion from a high-pressure chamber into a vacuum through an orifice produces internally cold, isolated gas phase atoms (or molecules). The high-pressure reservoir usually contains a dilute mixture of the desired molecule within a noble gas. The monoatomic noble gases have superior cooling properties due to the lack of internal degrees of freedom. If the ratio of stagnation pressure (high-pressure reservoir) to background pressure (vacuum) exceeds the value:

\[
\frac{P_0}{P_b} \geq \left[ \frac{(\gamma + 1)}{2} \right]^\frac{\gamma}{(\gamma - 1)},
\]

where \(\gamma\) is the ratio of heat capacities \(\gamma = C_p/C_v\) of the mixed gas (seed gas and sample), the velocity of the exiting beam will exceed the speed of sound in the gas mixture and undergo further expansion within the vacuum region \[115\]. This expansion can be approximated as adiabatic and isentropic, so most of the work done by the differential pressure modifies the internal energy of the gas. In the case of monatomic species, \(\gamma = 5/3\), the change of internal energy is a direct change on their kinetic energy. The expansion causes cooling of the expanding gas (narrowing of the Maxwell-Boltzman distribution) due to the large number of binary collisions. The expanding zone is insulated from the background gas by the shock wave barrier formed on the outskirts of the expanding envelope. The cooling zone is called the zone of silence and the speed of molecules is supersonic (Mach > 1). A cold collimated beam can be extracted from this expanding cloud with the use of a skimmer (a conical or parabolic differential pumping orifice). With this technique, temperatures as low as \(~1\) K are readily achieved.
4.1. Supersonic beams

The speed distribution of a supersonic beam can still be approximated by a Maxwell-Boltzman distribution with the central velocity shifted by $v_{\text{mean}}$:

$$v_{\text{mean}} = \sqrt{\frac{2}{\gamma - 1}} \frac{RT_0}{m} \left( 1 - \frac{P_b}{P_0} \right)^{\frac{\gamma - 1}{\gamma}},$$

(4.2)

where $R$ is the ideal gas constant, $m$ is the mass of the expanding atoms and $T_0$ is the temperature of the gas reservoir. With this equation and the Maxwell-Boltzman distribution it is possible to calculate the beam characteristics of various monoatomic gases with a reservoir at 300 K (see Table 4.1).

Table 4.1: Supersonic beam characteristics for noble gases with a reservoir temperature of 300 K.

<table>
<thead>
<tr>
<th>Species</th>
<th>Mean velocity (m/sec)</th>
<th>Velocity Spread ($\sigma_v$) (m/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>1800</td>
<td>22</td>
</tr>
<tr>
<td>Ne</td>
<td>900</td>
<td>11</td>
</tr>
<tr>
<td>Ar</td>
<td>600</td>
<td>17</td>
</tr>
<tr>
<td>Kr</td>
<td>450</td>
<td>11</td>
</tr>
<tr>
<td>Xe</td>
<td>390</td>
<td>9.8</td>
</tr>
</tbody>
</table>

With the use of pulsed valves (to limit the beam temporally and reduce pumping requirements), skimmers, and differential pumping, it is possible to make a dense collimated beam of stable species. If the desired species is metastable, or a free radical, the technique has to be modified so the species of interest is introduced at a point in the beam formation where any reactions are reduced to a minimum. This is usually done during the expansion.
4.2 Beam implementation

The hydrogen beam implementation had two purposes. The first purpose, which is the main topic of this dissertation, is to confirm that the Lyman-$\alpha$ source could detect cold hydrogen via Doppler-free LIF. This is the main discussion in this chapter. The second purpose is to perform hydrogen collision scattering experiments with ultra-cold Rb atoms trapped in a MOT. This particular collision experiment has not been performed by any other group to date. The collision experiments (H - Rb) are still ongoing and will not be discussed in this chapter. However, collisions of a beam of noble gases with the Rb-MOT were performed as a way of characterizing the beam; this is included here. There is a larger picture to this experiment that needs to be mentioned since a lot of the experimental equipment implementation only makes sense in view of this larger picture.

In addition to performing the LIF spectroscopy of hydrogen, this experiment is also setting the framework necessary for the photo-association of the RbH molecule. Theoretical predictions indicate that RbH has an enormous dipole moment. The dipole moment calculated is the largest of all the alkali-metal hydrides, which in turn is much larger than all the bialkalis. The dipole moment of RbH in the singlet state was calculated at 10.46 Debye (Competing only with CsH) \[116, 117\]. The large dipole moment of RbH can potentially lend itself to be used for electrostatic trapping and further cooling.

There is a large interest in ultra-cold polar molecules with large electric dipole moments. Among their applications are: the search for a permanent electric dipole moment of the electron \[118, 119\], quantum computation \[120\], creation of a dipolar superfluid \[121\] and a model system for lattice spin models \[122\]. However, the rich internal structure of molecules, which makes it difficult to implement a closed optical cycling transition, has prevented the realization of a MOT for molecules. An alternative to cooling down molecules directly is to produce them from ultra-cold atoms via photo-association. This was proposed in 1987 \[123\] and the year after, ultra-cold molecular ions were produced via associative photo-ionization.
4.2. Beam implementation

[124]. Since then, the quality of experimental spectroscopic data has been ahead of the ab-initio calculations pushing for refinement of theoretical models. All of this work has only been possible in diatomic molecules. The quest now is for ultra-cold heteronuclear molecules. The first ultra-cold heteronuclear molecule was $^6\text{Li}^7\text{Li}$ [125]. Heteronuclear molecules with states that support dipole moments of the order of Debye were first reported in 2004 [126].

To perform the photo-association, cold hydrogen, in addition to ultra-cold Rb, is required. For these reasons the beam implementation incorporate various additional features such as a Zeeman decelerator and a Zeeman bender. The purpose of the implementation is to deliver a beam of cold hydrogen atoms (with tuneable kinetic energy) for trapping or collision scattering and photo-association experiments. Fig. [4.1] shows a schematic of the hydrogen beam main components.

The experimental set up consists of the following modules:

- Source chamber where the hydrogen beam is created by entrainment (pick up) of hydrogen by a supersonically expanding beam of a noble gas.

- Zeeman decelerator chamber where the hydrogen atoms in a Low Field Seeking (LFS) state is decelerated (not discussed in this dissertation).

- Zeeman bender or filter chamber where the desired population of LFS hydrogen atoms are separated from the ensemble of noble gas and fast hydrogen atoms.

Each chamber is differentially pumped and, in that way, we can reach a vacuum of $\sim 10^{-8}$ Torr after the Zeeman bender, with the beam on, while the source chamber pressure can be as high as $10^{-4}$ Torr, during the valve opening. Fig. [4.2] is a solidworks schematic of the complete vacuum setup.

The schematic also shows some additional features such as a magnetic focuser. This focuser is necessary if a well collimated beam is required after
4.2. Beam implementation

Figure 4.1: The experimental set up consists of the following modules: Source chamber where the hydrogen beam is created by entrainment (pick up) of hydrogen by a supersonically expanding beam of a noble gas. Zeeman decelerator chamber (not discussed in this thesis) where the hydrogen atoms in a Low Field Seeking (LFS) state are decelerated (cooled). Zeeman bender or filter chamber where the desired population of LFS hydrogen atoms is separated from the ensemble of noble gas and fast hydrogen atoms.

the bender as the hydrogen atoms exiting the bender have large transversal velocity and spread out. Also the Lyman-α cell can be attached to the system in two different positions; at the source chamber, for LIF of the hot hydrogen, and at the end of the Zeeman decelerator chamber, for Doppler-free LIF of the hydrogen beam. Detectors have been integrated such as a PMT, directly above the crossing point of the Lyman-α source and hydrogen beam, and a Quadrupole Mass Spectrometer (QMS), after the focuser. The QMS can also be placed on the Zeeman chamber instead of the PMT. Fig. 4.3 shows some pictures of each item.

Regarding this dissertation, two sections of the experimental set up are important. The source chamber where the hydrogen beam is generated and the Zeeman bender which is used to confirm the hydrogen in the beam.
Figure 4.2: CAD drawing of the complete vacuum setup. The Lyman-α cell can be placed in the source chamber for spectroscopy of hot hydrogen or in the Zeeman decelerator chamber for Doppler free spectroscopy of the beam.
Figure 4.3: Pictures of the main components: the pulsed valve, Zeeman decelerator and Zeeman bender. The focuser at the end of the decelerator was not used for these experiments.

4.2.1 Source chamber

To create the hydrogen beam we have used a home-built pulsed valve actuated by a solenoid. The valve releases high pressure gas (seed gas) into the vacuum creating a supersonic expansion. Hydrogen gas (atomic hydrogen) is generated by the thermal cracker which is placed perpendicular to the beam and directly in front of the valve. As the gas is expanding some hydrogen atoms go through the shock barrier and make it into the zone of silence. Here they are “picked-up” by the expanding gas and cooled due to the large number of binary collisions with the seed gas. The beam is extracted from the expansion with a parabolic skimmer where it goes into a high vacuum chamber. The source chamber has a large gas load from the gas cracker and the pulse valve, and it is pumped down with a 1000
4.2. Beam implementation

L/sec turbomolecular pump. The pressure during valve opening is as high as $10^{-4}$ Torr.

Pulsed valve

The design of the pulsed valve is based on the Even Lavie design [127] and was developed by Pavle Djuricanin (mechanical) and Sajjad Haidar (electronics) from Technical Services in the Chemistry Department. Fig. 4.4 shows the main valve components. The valve consists of a spring loaded plunger that seals the nozzle opening. During actuation a high current pulse drives a solenoid around the plunger. The force created by the magnetic field pushes the plunger against the spring. The coil of the solenoid uses a Permendure shield as field concentrator. The nozzle has a conical shape with an opening angle of 40 deg. According to the literature this is the most optimal configuration to produce low spread (0.43 Steradians) directional beams [128]. The nozzle has a pinhole of 250 μm diameter. The main problem with this configuration is that small misalignments between the nozzle and the skimmer result in dramatic reductions of the beam intensity. This is very critical for this setup as the skimmer has been placed at a long distance from the valve in order to minimize the transversal velocity of the beam (minimizing any Doppler broadening during LIF experiments). The plunger and solenoid design were optimized to maximize the opening stroke force (and speed) by simulating the magnetic field and force at different configurations.

The force generated by the solenoid can be calculated with the Maxwell stress tensor:

$$F = \frac{1}{\mu_0} \oint_A \left( \vec{B} (\vec{B} \cdot \vec{n}) - \frac{1}{2} \vec{B}^2 \cdot \vec{n} \right) dA,$$

(4.3)

where $\vec{n}$ is normal to the surface area A. The magnetic field was calculated at every point for various positions of the solenoid and the most optimal position is to have the centre of the air gap in the close position (surface A shown in Fig. 4.5) offset from the middle of the solenoid by 750 μm. Fig. 4.5 shows the result of the simulations (the best configuration). The valve opening times are as small as 20 μsec with repetition rate as high as 1 kHz.
4.2. Beam implementation

The design of the pulsed valve is based on Even Lavie design [127] and was developed by Technical Services in the Chemistry Department. The valve consists of a spring-loaded plunger that seals the nozzle opening with a reciprocating (up-and-down) motion. During actuation, a high current pulse drives a solenoid around the plunger. The force created by the magnetic field pushes the plunger against the spring. The coil of the solenoid uses a Permendure shield as a field concentrator. The nozzle has a conical shape with an opening angle of 40 deg. According to the literature, this is the most optimal configuration to produce low spread (0.43 Steradians) directional beams [128]. The nozzle has a pinhole 250 µm diameter.

Figure 4.4: The design of the pulsed valve is based on Even Lavie design [127] and was developed by Technical Services in the Chemistry Department. The valve consists of a spring-loaded plunger that seals the nozzle opening with a reciprocating (up-and-down) motion. During actuation, a high current pulse drives a solenoid around the plunger. The force created by the magnetic field pushes the plunger against the spring. The coil of the solenoid uses a Permendure shield as a field concentrator. The nozzle has a conical shape with an opening angle of 40 deg. According to the literature, this is the most optimal configuration to produce low spread (0.43 Steradians) directional beams [128]. The nozzle has a pinhole 250 µm diameter.
and stagnation pressures as high as 300 psi.

To monitor the gas pulse in the chamber two discharge plates were placed in front of the valve nozzle. By applying a high voltage across the gas during the expansion it is possible to generate plasma. The ions and electrons accelerate to the plates and a current signal proportional to the gas plasma density can be measured. Fig. 4.6 shows the profile of the gas pulse during the valve opening. Pulse widths as short as 30 µsec can be generated. By monitoring the current it is possible to confirm the opening time and also that the valve is not bouncing and opening multiple times. The bouncing occurs when the current supplied to the solenoid is too high and the plunger goes beyond some equilibrium point. The optimal current is a function of the gas pressure and life time of the spring. Before performing experiments the discharge plates are used to optimize the current to the valve until there is no bouncing. Also the delay time and opening time can be read from the current profile.

**Hydrogen entrainment**

For the LIF spectroscopy the expanding gas used (seed gas) was Ar. The hydrogen was sourced from the gas cracker in front of the expansion. To increase the density of the hydrogen (and increase the probability of hydrogen atoms penetrating the zone of silence) an entrainment (pick up) cell was placed in front of the expanding beam. Fig. 4.7 shows the configuration of the valve, cracker, entrainment cell and skimmer. Also a schematic of the pick up cell and mechanism is shown. The entrainment cell was made of 1/4 inch quartz tubing and connected to the gas cracker. The main limitation of using an entrainment cell is that recombination of rarified hydrogen occurs predominantly on the available surfaces. Another option to reduce the recombination is to place the hydrogen effusive source very close in front of the expanding beam without an entrainment cell. The two options were tested but no significant difference was observed. However, by optimizing the geometry of the entrainment cell and possibly coating the surface with Teflon™ it may be possible to get a larger percentage of
Figure 4.5: The pulsed valve stroke was optimized by maximizing the force exerted by the solenoid on the plunger. The force generated by the solenoid can be calculated with the Maxwell stress tensor (Eq. 4.3). The integration is done on the surface A. The magnetic field calculation for the best configuration is shown.
Figure 4.6: To monitor the gas pulse in the chamber two discharge plates were placed in front of the valve nozzle. By applying a high voltage across the gas during the expansion it is possible to ionize it and generate plasma. The ions and electrons accelerate to the plates and a current signal proportional to the gas plasma density can be measured. The flat top of the plasma profile indicates that the valve is fully open during the pulse. The second profile indicates that the plunger is bouncing. The bouncing can be minimized by reducing the driving current.
4.2. Beam implementation

Figure 4.7: To generate the hydrogen beam the hydrogen was sourced from gas cracker in front of the expansion. To increase the density of the hydrogen (and increase the probability of hydrogen atoms penetrating the zone of silence) an entrainment (pick up) cell was placed in front of the expanding beam. The expanding (seed) gas was Ar.

hydrogen in the beam with this technique. Due to the large recombination factor a large percentage of the hydrogen picked up by the beam is molecular hydrogen. An advantage of using this technique in contrast to photodissociation (a well known technique) is that the the flow of atomic hydrogen is continuous. The gas cracker can continuously dissociate up to 90% of 1 sccm of H$_2$. Photodissociation usually require nanosecond long pulse lasers. With the gas cracker technique hydrogen can be entrained into an expanding beam that is temporarily limited by the valve and pumping speed of the system. Our system can potentially generate pulses up to 200 µsec. For applications such as photo-association that are not time
4.2. Beam implementation

constrained the gas cracker technique can potentially deliver much more hydrogen atoms than photoassociation.

4.2.2 Zeeman bender

In order to filter the beam from unwanted atoms (such as the Ar seed gas) and molecules (H$_2$) a filter has been implemented. The filter is a magnetic quadrupole bender that uses the Zeeman effect to guide the LFS atoms to the collision chamber while dispersing everything else. When hydrogen is placed in a magnetic field the degeneracy of the $F = 1$ states is broken into the three $M_F = 0, \pm 1$, states by the Zeeman hamiltonian (see Fig. [1.1]):

$$H_z = \mu^e s_z B + \mu^p I_z B,$$

(4.4)

where $\mu^p$ and $\mu^e$ are the magnetic moment of the electron and proton, $s_z$ and $I_z$ are the spins and $\vec{B} = (0, 0, B)$. The energy of the $M = 0$ and 1 states increases with magnetic field. These states are called LFS’s as when there is a field gradient they feel a force opposite to the gradient (towards the lower fields):

$$F = -\mu \nabla \vec{B}.$$

(4.5)

The magnetic field is created by passing a current through each leg of the quadrupole. A magnetic minimum is created in the middle of the quadrupole by making each leg current anti-parallel to its nearest neighbours. A simulation of the magnetic field shows a magnetic minimum in the middle where the LFS atoms are guided (Fig. [4.8]). The force is proportional to the field gradient and to the current applied. It is possible to select the maximum speed of hydrogen atoms that would be guided by the quadrupole by adjusting the current. The maximum current for the quadrupole is 2000 Amps and limited only by the driving electronics. The other limitation is the transit time as the current pulse can only be sustained for 1.5 msec. However, at the speed of supersonically expanded Ar (the seed gas of the beam) this is not an issue. Two quadrupoles were implemented. The quadrupole used in this experimental set up is a 90 deg
4.2. Beam implementation

turn and 110 cm radius with a leg spacing of 2.36 mm. This quadrupole can bend hydrogen atoms at velocities of 580 m/sec when operated at 2000 Amps. The second quadrupole is a 10 deg and 8 m radius with a leg spacing of 2.36 mm. This quadrupole can bend hydrogen at velocities of 2500 m/sec when operated at 1500 Amps. Fig. 4.9 shows both quadrupoles.

The quadrupole was used to confirm that the beam contained hydrogen. As mentioned in the previous section the beam contained a large percentage of molecular hydrogen. To characterize the beam a QMS was used. The QMS uses high energy electrons (∼70 eV) to ionize the species. At this electron energy everything is ionized. Particles with the right mass to charge ratio are guided through the quadrupole towards the channeltron, where the current is directly measured from the anode. When molecular hydrogen is ionized 2% of it dissociatively ionizes into a H⁺ and is seen as mass 1 by the QMS. Since a large percentage of the beam and the background gas in the chamber is molecular hydrogen, it is very difficult to discern the hydrogen in the beam from the fraction of dissociated H₂. The Zeeman bender can only extract hydrogen from the beam and it has been used to confirm that the beam, in fact, contains hydrogen picked up during the expansion. This is described in the next section.
4.2. Beam implementation

Figure 4.8: The Zeeman bender was implemented with a quadrupole. The magnetic field is created by passing a current through each leg of the quadrupole. A magnetic minimum is created in the middle of the quadrupole by making each leg current anti-parallel to its nearest neighbours. A simulation of the magnetic field shows a magnetic minimum in the middle where the LFS atoms are guided. The force is proportional to the gradient of the magnetic field and to the current applied. It is possible to select the maximum speed of hydrogen atoms that would be guided by the quadrupole. The maximum current for the quadrupole is 2000 Amps and limited only by the driving electronics.
4.2. Beam implementation

Figure 4.9: Two quadrupoles where implemented. The quadrupole that was tested in this experimental set up is a 90 deg turn and 110 cm radius with a magnetic minimum size of 2 mm. This quadrupole can bend hydrogen atoms at velocities of 580 m/sec when operated at 2000 Amps. The second quadrupole is a 10 deg and 8 m radius with a magnetic minimum size of 2 mm. This quadrupole can bend hydrogen at velocities of 2500 m/sec when operated at 1500 Amps.
4.3 Hydrogen Lyman-α detection

The hydrogen beam implemented is composed of Ar (the seed gas), molecular hydrogen H₂ and atomic hydrogen. The molecular hydrogen is the result of the recombination that occurs during the entrainment process. The Ar seed gas expansion results in a beam of approximately 600 m/sec. In order to perform Doppler free LIF spectroscopy the skimmer was placed far from the valve to reduce the transverse velocity of the hydrogen. With a skimmer of 2 mm diameter and a skimmer to valve distance of 25 cm the collimation factor is 125. The transversal velocity of the hydrogen is less than 4.8 m/sec. This is equivalent to a transverse temperature of 0.0011 K and a Doppler broadening of 58.9 MHz. The total linewidth of the hydrogen transition can be expected to be ~ 150 MHz. The source linewidth was estimated to be 6 GHz so the ratio of the linewidths is 40:1. The source photons are much less efficient in inducing fluorescence. The low density of hydrogen and photons result in a very small fluorescence signal at the threshold of detection. To be able to see any signal the PMT gain had to be increased to the maximum (10⁵), but this can only be done by reducing the scattered ωR photons.

QMS of filtered hydrogen

Before attempting any spectroscopy the hydrogen in the beam was confirmed by measuring the amount of hydrogen after the Zeeman bender. This was done with the QMS, which filters the charge to mass ratio selected, by measuring the current signal directly from the channeltron. The QMS was initially calibrated by filling a chamber with up to ~ 10⁻⁵ Torr of He and comparing the current signal with a cold cathode gauge. The same calibration was done with molecular hydrogen. Calibration of hydrogen was done with molecular hydrogen since 2% of it ionizes into H⁺ [100]. With this calibration it was possible to estimate a number density after the bender. Fig. 4.10 shows the experiment with the bender. During the experiment the pulsed valve was running and the bender was switched on and off while the QMS was always reading the density of atomic hydrogen.
4.3. Hydrogen Lyman-$\alpha$ detection

Since the pressure at the QMS chamber was $\sim 10^8$ Torr, and most of the residual gas is H$_2$, the QMS signal had to be averaged over long periods. Fig. 4.10 shows the QMS signal in which a window average of 1000 has been applied. The valve and bender were operated at 5 Hz and the window average covers 3.33 minutes of data. When the molecular hydrogen flow into the gas cracker is decreased, the pressure in the source chamber and in the QMS chamber decreases. At a base pressure of $\sim 10^{-8}$ Torr and 60% H$_2$ there are $\sim 10^{10}$ H$_2$ molecules/cm$^3$ in the chamber. When ionized 2% of them dissociate into hydrogen ions which is $10^8$ ions/cm$^3$ in the ionizer volume. All the measurements performed are over this background. From the QMS signal it was estimated that $\sim 10^8$ hydrogen atoms/cm$^3$ over the background were added when the bender was turned on. The valve opening was 50 µsec and the beam diameter is 2 mm. The beam has a mean speed of 600 m/sec so every pulse is approximately 0.94 cm$^3$. From this we can estimate $\sim 10^8$ hydrogen atoms/pulse being guided.
4.3. Hydrogen Lyman-α detection

Figure 4.10: The hydrogen in the beam was confirmed by measuring the amount of hydrogen after the Zeeman bender. This was done with the QMS by measuring the current signal directly from the channeltron amplifier. The QMS was initially calibrated by filling a chamber with He and comparing the current signal with a cold cathode gauge. The same calibration was done with molecular hydrogen. Calibration of hydrogen was done with molecular hydrogen since 2% of it ionizes into H$^+$ [100]. During the experiment the pulsed valve was running and the bender was switched on and off while the QMS was always reading the amount of hydrogen. Since the pressure at the QMS chamber was $\sim 10^{-8}$ Torr, and most of the residual gas is the H$_2$, the signal, the QMS signal had to be averaged over long periods.
4.3. Hydrogen Lyman-α detection

QMS Time of flight

To verify the beam time of flight, the QMS was placed on the PMT port (in front of the skimmer). When the QMS is running continuously, it is possible to see the beam profile (provided there is enough signal to noise ratio). When looking for mass 1 particles the time of flight profile captured by the QMS is mostly due to hydrogen molecules. However, a small percentage is due to the hydrogen atoms. To discern the hydrogen atoms from the background hydrogen the beam was averaged 128 times and the difference between a beam with the gas cracker power switched on and switched off was taken (the molecular hydrogen is continuously flowing). Fig. 4.11 shows the time of flight profile and the difference of the beams which is attributed to hydrogen atoms. From the time of flight profile the number of hydrogen atoms per pulse is estimated to be $2.5 \times 10^{10}$ atoms/pulse. This means that the bender can bend approximately 1% of atoms. An interesting feature of the profile is that it appears to be two regions of higher density of hydrogen around the main body of the pulse. This is consistent with reports of beam separation when the seed gas is a heavier atom [129]. In this case, Ar with mass 40, appears to push the hydrogen atoms in front and behind the pulse.
4.3. Hydrogen Lyman-α detection

Figure 4.11: To verify the beam the QMS was placed on the PMT port. When the QMS is running continuously it is possible to see the beam profile (provided there is enough signal to noise ratio). When looking for mass 1 particles the time of flight profile capture by the QMS is mostly due to hydrogen molecules. However, a small percentage is due to the hydrogen atoms. To discern the hydrogen atoms from the background hydrogen the beam was averaged 128 times and the difference between a beam with the gas cracker on and off was taken. From the time of flight profile the number of hydrogen atoms per pulse is estimated to be $2.5 \times 10^{10}$ atoms/pulse. This means that the bender can bend approximately 1% of atoms. An interesting feature of the profile is that it appears to be two regions of higher density of hydrogen around the main body of the pulse.
4.3. Hydrogen Lyman-α detection

Fluorescence experiment

In Chapter 3 the density of hydrogen in the spectroscopy chamber was calculated to be \( \sim 10^{14} \) hydrogen atoms/cm\(^3\). The density of hydrogen in the beam is estimated to be 4 orders of magnitude less. Furthermore, the linewidth ratios of the transition and the source indicate that the photons are at least 40 times less efficient at inducing fluorescence. To perform the LIF experiment the chamber was modified to reduce the scattered \( \omega_R \) photons and to increase the solid angle view of the detector. Blackened baffles, to reduce the scattered light, and collimating lenses, to capture more fluorescence photons into the PMT, where placed around the interaction area. All the lenses used were MgF\(_2\). Fig. 4.12 shows the setup. The diameter of the beam is 2 mm and the radius of the laser beam at the focal point is less than 10 \( \mu \)m. The flashlight and Q-switch of the pump laser were controlled by a computer that also set the timing for the valve. The timing for the valve opening was scanned and at every time step the signal was recorded and averaged multiple times. The timing for the valve opening was scanned in order to find the point with the highest hydrogen density along the beam. The main difficulty performing this experiment was the laser stability. The laser is not stable enough for long runs and the power of the Lyman-α fluctuates. Power fluctuations are due to temperature changes in the room and in the cooling water and also due to misalignment of optical components due to vibration. Averaging is required since the PMT is operating very close to the detection threshold and the signal to noise ratio is very small. Fig. 4.13 shows the results of the scans. According to the QMS time of flight signal the hydrogen pulse extends from 0.5 to 1.2 msec after the valve opening with a maximum hydrogen density at 1.2 msec. The maximum LIF signal is just between 1.2 and 1.3 msec and this agrees with the QMS. However, the LIF signal does not appear to reflect the density profile of the beam and this is troubling. The two data samples shown are the only LIF signal that was obtained after several experiments and it was not possible to improve it. The signal, however, shows a clear signature of LIF at the time that it is expected and it is always clearly distinguishable.
4.3. Hydrogen Lyman-α detection

from noise. One potential problem is the alignment/overlap of the laser and the hydrogen beam. As currently designed adjustments of the beam and laser are very difficult to perform and for that reason it was not possible to further optimize the experiment. Furthermore, since the generation of Lyman-α radiation was already confirmed (see Chapter 3) and these LIF experiments showed some signatures of hydrogen fluorescence it was decided that continuing this experiment was not necessary.

Figure 4.12: To perform the LIF experiment the chamber was modified to reduce the scattered $\omega_R$ photons and to increase the solid angle view of the detector. Blackened baffles to reduce the scattered light and collimating lenses where placed around the interaction area to capture more fluorescence photons into the PMT. All the lenses used were MgF$_2$. 

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4.3. Hydrogen Lyman-α detection

Figure 4.13: The flashlight and Q-switch of the pump laser were controlled by a computer that also set the timing for the valve. The laser timing was scanned to find the highest density of hydrogen along the profile of the beam and every time the signal was averaged multiple times. The difficulty with this experiment was that the laser is not stable enough for long runs and the power of the Lyman-α fluctuates. Stability can only be accomplished for short amounts of time and the experiment runs need to be short. Averaging is required since the PMT is operating very close to the detection threshold and the signal to noise ratio is large.
4.4 Collision scattering experiments

It has been proposed that the measurement of background gas collisions with a cloud of trapped atoms can be used as a standard of the background gas density [130–132]. This measuring technique (as will be shown in this section) reduces to the calculation of a scattering cross-section. The measurement does not require any calibration (in contrast to ionization gauges or other techniques) and for that reason it could be used as a measurement standard. In a MOT the rate of change of the number of atoms in the trap can be given as:

\[
\frac{dN}{dt} = R - \Gamma N - \beta \int n_{\text{MOT}}^2(r,t)d^3r.
\]

(4.6)

Here R is the rate of atom capture, \(\Gamma N\) is the rate of loss due to collisions of the trapped atoms with the background gas atoms and \(\beta\) is the rate of loss due to collisions of two trapped atoms whose density is \(n_{\text{MOT}}\). If the density is taken to be constant then:

\[
N(t) = \frac{R}{\Gamma_{\text{eff}}} \left(1 - \exp\{-\Gamma_{\text{eff}}t\}\right),
\]

(4.7)

where \(\Gamma_{\text{eff}} = \Gamma + \beta n_{\text{MOT}}\). In a magnetic trap R is zero and if \(\beta n_{\text{MOT}} << \Gamma\) then:

\[
N(t) = N(0) \exp\{-\Gamma t\}.
\]

(4.8)

In this case the rate of change of the number of atoms is only a function of the loss-rate constant:

\[
\Gamma = n_{\text{Bkg}} \langle \sigma_T(v)v \rangle,
\]

(4.9)

where \(\langle \sigma_T(v)v \rangle\) denotes an average over the centre of mass speed \(v\) of the colliding atoms, and \(\sigma_T\) is the collisional cross section of the trapped atoms with the background atoms of density \(n_{\text{Bkg}}\). If the velocity-averaged collision cross section is known, then by measuring the rate of loss of the trapped atoms it is possible to deduce \(n_{\text{Bkg}}\), the density of background gas. This measurement does not rely on any other calibration method and it can
provide a primary standard for density measurements. Since the density measurement provided by a MOT is localized, this method can be used to measure density differences in a volume with a resolution given by the MOT size. For instance, the density of an atom beam can be measured with this technique.

There are two main differences between a background gas density measurement and a beam density measurement: the type of velocity-average performed and the time scales involved. In the case of background gas the average is over a Maxwell-Boltzman distribution with a mean speed given by the temperature of the ensemble. In the case of a beam, the distribution mean is the supersonic speed given mainly by the stagnation pressure and temperature of the reservoir. Regarding the time scales, the rate of change of background gas is usually slow compared with the time between collision events. In fact $\Gamma$ is assumed to be time independent. That is certainly not the case in a beam density measurement where the density changes in the order of hundreds of microseconds.

In a beam density measurement the loss-rate constant has a component due to background collisions and a component due to the collision with the beam; and if $\beta n_{\text{MOT}} \ll \Gamma$ then:

$$\frac{dN}{dt} = R - (\Gamma_{\text{Bkg}} + \Gamma_{\text{Beam}}(t))N. \quad (4.10)$$

In the collision experiments that have been performed the MOT is always in equilibrium, $R - \Gamma_{\text{Bkg}}N = 0$, before the collision. The time scale of the beam collision event is less than one millisecond and the change in the number of atoms trapped is less than 1%. It is reasonable to assume that:

$$\frac{dN(t)}{dt} \approx -N(t)\Gamma_{\text{Beam}}(t). \quad (4.11)$$

Fig. 4.14 is a plot of the scattered light from a MOT as a function of time. It shows a collision event where a beam of Ar collided with a Rb-MOT. The negligible rate of change before and after the collision support the previous assumptions. From the scattered light signal is possible to extract
4.4. Collision scattering experiments

\[ \Gamma_{\text{Beam}}(t) = n_{\text{Beam}}(t) (\sigma_{\text{MOT-Beam}}(v)v). \]  

Here \( v \) is effectively \( v_{\text{Beam}} \) since the MOT atoms can be assumed to be stationary compared to the supersonic speed of the beam. In this section it will be shown that in principle, this signal has all the information required to characterize the beam.

![Diagram of collision scattering experiments](image)

Figure 4.14: Scattered light from a MOT as a function of time. It shows a collision event where a beam of Ar collided with a Rb-MOT.

To perform the collision experiments, a \(^{85}\text{Rb} \) MOT was implemented and integrated with the supersonic beam. The trapping technique uses the D2 Rb transitions: \((4p^5(2P^o_{3/2})5p^2)[3/2] \leftarrow 4p^5(2P^o_{1/2})5s^2[1/2]\)\(\), 138794.380 cm\(^{-1}\) - 158151.666 cm\(^{-1}\), from the \(F = 3\) to the \(F = 4'\) hyperfine levels (the “pump”). To close the cycle (since some transitions occur to the \(F= 2\) level) a “re-pump” transition is induced from the \(F = 2\) to the \(F = 3'\) levels. MOTs
4.4. Collision scattering experiments

...rely on the momentum imparted by individual photons for trapping and cooling. To select the atoms to which momentum must be imparted, lasers are detuned from the resonance. The balance between Doppler cooling forces associated with photon absorption and the subsequent heating due to re-emission of absorbed photons leads to the so-called Doppler cooling limit. The Doppler cooling limit for $^{85}\text{Rb}$ is 145.57 $\mu\text{K}$. The light for the MOT has been transported from Dr. Kirk Madison’s laboratory via a polarization maintaining fiber which carries both the pump and the perpendicularly polarized re-pump. This particular MOT was implemented in a glass tube (square stock) bonded to Conflat™ flanges. In addition to the anti-Helmoltz coils a couple of compensation coils have been added to allow steering of the MOT in the $xy$ plane. The MOT chamber is pumped with an ion pump at the beam entrance and a turbo pump at the exit. Two Rb wire evaporation dispensers have been used: Alvasource and SAES Advanced Technologies. By passing some current through the wire the Rb metal is evaporated. Fig. 4.15 shows an schematic of the experimental set up.

To integrate the MOT with the beam the Zeeman bender has been removed and the MOT chamber has been attached directly in front of the beam. To perform the measurements the background density has to be stable at the MOT chamber during valve opening. This has been accomplished by adding a second skimmer before the MOT and a turbomolecular pump behind it. With the additional differential pumping stage the pressure at the MOT is $\sim 10^{-8} - 10^{-9}$ Torr. The turbomolecular pump after the MOT captures the beam after the collisions. The skimmer, when properly aligned with the valve, first skimmer and MOT, does not affect the beam as its opening diameter is bigger than the first skimmer. Fig. 4.15 shows some pictures of the components.
4.4 Collision scattering experiments

Figure 4.15: To perform the collision experiments a MOT chamber has been attached directly in front of the beam. To perform the measurements the background density has to be stable at the MOT chamber during valve opening. This has been accomplished by adding a second skimmer before the MOT and a turbomolecular pump behind it. With the additional differential pumping stage the pressure at the MOT is $\sim 10^{-8} - 10^{-9}$ Torr. The turbomolecular pump after the MOT captures the beam after the collisions.
4.4. Collision scattering experiments

To measure the MOT atom number $N(t)$ a photodiode is used. The photodiode signal is proportional to the MOT number:

$$V_{ph}(t) = \alpha N(t) - V_{N=0}. \quad (4.12)$$

The constant $\alpha$ is a function of the photodiode collection efficiency and the scattering rate of photons by the atoms in the trap. As long as the photodiode is operated in a linear region, $\alpha$ can be assumed to be constant. The bias signal $V_{N=0}$ is the product of the laser’s photons scattering from the walls of the glass tube and leaked photons from the ambient light. To measure the density of the beam all that is needed is the photodiode signal:

$$\frac{dN(t)}{dt} \approx -N(t)\Gamma_{\text{Beam}}(t) \Rightarrow \frac{V_{ph}(t)}{dt} \approx -(V_{ph}(t) - V_{N=0})n_{\text{Beam}}(t)\langle \sigma v_{\text{Beam}} \rangle. \quad (4.13)$$

The cross section $\sigma_{\text{MOT–Beam}}(v_{\text{Beam}})$ can be calculated from first principles under some assumptions of the inter-atomic potential. The speed distribution of the beam $f(v_{\text{Beam}})$ can be extracted from the normalized density signal:

$$-\frac{1}{\max(n_{\text{Beam}})} \left[ n_{\text{Beam}}(t) \approx \frac{V_{ph}(t)}{dt} \frac{1}{(V_{ph}(t) - V_{N=0})\langle \sigma v_{\text{Beam}} \rangle} \right]. \quad (4.14)$$

However, there is some time delay caused by the transit time of the MOT atoms. These atoms have to move out of the detection area of the photodiode. This can be optimized by narrowing the photodiode view area, but the transit time represents the ultimate time resolution possible with this measuring technique. Nevertheless, with this information the constant $\langle \sigma_{\text{MOT–Beam}}(v_{\text{Beam}}) \rangle$ can be calculated by proper averaging:

$$\langle \sigma_{\text{MOT–Beam}}(v_{\text{Beam}}) \rangle = \frac{\int_{v=0}^{v=\infty} \sigma_{\text{MOT–Beam}}(v_{\text{Beam}})v_{\text{Beam}}f(v_{\text{Beam}})dv_{\text{Beam}}}{\int_{v=0}^{v=\infty} f(v_{\text{Beam}})dv_{\text{Beam}}}.$$ \hspace{1cm} (4.15)

Finally, the calculation of $\sigma_{\text{MOT–Beam}}$ is essentially a calculation of the scat-
tered spherical wavefunction \( g(\nu_{Beam}, \Omega) \), where \( \Omega \) is the solid angle:

\[
\sigma_{MOT_{Beam}(\nu_{Beam})} = \int |g(\nu_{Beam}, \Omega)|^2 \, d\Omega. \tag{4.16}
\]

The potential \( U(r) \) used in the Schrödinger equation to calculate the scattered wavefunction is assumed to be of the Lennard-Jones type (this is the assumption mentioned before):

\[
U(r) = U_{L,J}(r) = \frac{C_{12}}{r^{12}} - \frac{C_6}{r^6}. \tag{4.17}
\]

The sixth-power term in the Lennard-Jones potential arises from instantaneous dipole-induced dipole interactions (London dispersions forces), which represent interactions in noble gases very well. The twelfth-power term has no physical basis and it is chosen for convenience of calculation (it is the square of the attractive term). However, for large potential wells (or low kinetic energy), where attractive forces dominate the interatomic interactions, the \( C_6/r^6 \) term dominates, and this is the most common case.

In addition, according to classical calculations of gas kinetics during a supersonic expansion the reduced collision cross section \( \sigma^* \) (in units of \( \pi r_m^2 \), where \( r_m \) is the equilibrium potential) can be calculated as [133, 134]:

\[
\sigma^* = 1.48K^{-1/3} \quad \Rightarrow \quad K = \frac{\mu v^2}{2\epsilon}, \tag{4.18}
\]

where \( \mu \) is the reduced mass and \( \epsilon \) is the potential well. This approximation can be used as long as \( K < 2 \) and it is good for most gases where the stagnation temperature is around, or below, room temperature. For very high reduced energies (\( K > 5 \)), the cross section becomes \( \sigma^* = 0.74K^{-1/6} \). This classical calculation gives some insight into the collision experiment and can be used to approximate a cross section. For instance, the atom loss rate coefficient has been calculated from these classical approximations and for the Ar/ Rb system \( \Gamma/ P \) (P as the pressure of Ar) is \( 2.3 \times 10^7 \text{ Torr}^{-1} \text{ sec}^{-1} \) [131]. This calculation assumes a 1 K trap depth and 300 K. Measurements with similar trap depths obtained \( 2.3 \times 10^7 \text{ Torr}^{-1} \text{ sec}^{-1} \) [135].
4.4. Collision scattering experiments

In summary, the term $\langle \sigma_{\text{MOT-Beam}}(v_{\text{Beam}})v_{\text{Beam}} \rangle$ can be calculated and it is only a constant in this measurement. In fact, this term has been calculated for Ar. This calculation, however, is not the author’s work and details are not presented in this dissertation. Regarding this discussion, it is reasonable to assume that the term $\langle \sigma_{\text{MOT-Beam}}(v_{\text{Beam}})v_{\text{Beam}} \rangle \approx 10^{-10} \text{ cm}^3/\text{sec}$ [136]. Fig. 4.16 shows the photodiode signal from measurements of collisions of three gases: Ar, Kr and He. The signal is quite noisy but this is not an inherit limitation of the technique and it can be improved by hardware changes. The data has been smoothed using a windowed average. The second plot on the right is the averaged signal and its derivative. The derivative is proportional to the density of the beam $n(t)$. The QMS signal is also shown for comparison. An interesting feature is what appears to be two different peaks in the Kr density. The QMS signal also shows these feature but it is less pronounced. Perhaps, the beam separated or the valve bounced, opening a second time. From the derivative of the signal the velocity distribution can be extracted. The absolute number of the density can be calculated by multiplying the constant $\langle \sigma_{\text{MOT-Beam}}(v_{\text{Beam}})v_{\text{Beam}} \rangle$. Fig. 4.17 shows the density and absolute atom number and a fit of the velocity distribution.
4.4. Collision scattering experiments

Figure 4.16: Photodiode signal for three different experiments with Ar, Kr and He. The signal has been averaged and the derivative is shown on the right. The QMS signal is also shown for comparison. A particular feature is the what appears to be two different beams in the Kr density. The QMS signal also shows these feature but it is less pronounced.
4.4. Collision scattering experiments

![Graphs of density over time for Ar, Kr, and He gases with corresponding speed distribution fits and mean speeds.](image)

Figure 4.17: From the derivative of the signal the velocity distribution can be extracted. The absolute number of the density can be calculated by multiplying the constant \( \langle \sigma_{MOT-Beam} \rangle \cdot \nu_{Beam} \nu_{Beam} \).
4.4. Collision scattering experiments

In conclusion, a MOT can be used as a standard to measure the absolute gas density, and more importantly, to measure very localized atomic densities. The beam used to demonstrate the measurement technique has a diameter of 2 mm. The MOT diameter is $\sim 1-2$ mm diameter in size and this is the volume resolution of the measurement. The time resolution is given by the time it takes the trapped atoms to escape the photodiode view area. In the experiment we can steer the MOT up to 1 cm in the $xy$ plane by using compensation coils. By moving the MOT 2.5 mm off the beam the signal disappears and this shows the potential of the technique. If the detection can be improved, MOTs as small as a few hundred micrometers can be used.

As initially stated, the purpose of the hydrogen beam development is to eventually perform the photo-association of the RbH molecules. Hydrogen collision experiments have also been performed to this end. However, without the Zeeman bender, the beam is a mixture of hydrogen molecules, hydrogen atoms and some seed gas. The hydrogen collision experiment was performed but the analysis is more difficult with the mixture of species. In fact, to perform the analysis, additional information of the species abundance is needed. The bender was not used because the density of the hydrogen atoms is too low to be observed with the current system. Nevertheless, the main purpose of this exposition is to show the potential of the technique. To measure the scattering of Rb atoms other techniques can be used such as “hot wire” (Langmuir-Taylor detector). However, the purpose of this section is to demonstrate the MOT potential as a measuring device. Regarding the RbH experiments the current limitation is the hydrogen source. The experiment requires a beam of very slow and high density hydrogen. In order to increase the hydrogen density, different sources are being explored and this is discussed in the next chapter. Also the current implementation of the Lyman-α is discussed.
Chapter 5

Current work and conclusion

In this chapter the current developments regarding the Lyman-\(\alpha\) source and the hydrogen source are discussed. The final form of the Lyman-\(\alpha\) source is being developed. This includes the laser beam delivery system at the CERN facility and the equipment required for optical cooling and spectroscopy of antihydrogen. Regarding the hydrogen beam and scattering experiments the main focus is on increasing the density of hydrogen in the beam. Although the beam as implemented delivers sufficient hydrogen to perform mass spectroscopy, a higher number of hydrogen atoms is required to observe photo-association with the Rb MOT.

5.1 Lyman-\(\alpha\) Source

The RE-FWM process is very efficient and can potentially produce a very high yield of VUV photons. However, the system is very complex and it is not very stable. Experiments at ALPHA require a very stable and robust system. It was decided that the THG process would be a better candidate. The input laser required for the THG can be implemented with solid state lasers. This type of system is more robust and requires less maintenance than a dye laser based system. Furthermore, to perform cooling, a narrow bandwidth laser of \(\sim 100\) MHz is required, and this is easier to accomplish with solid state systems. Another limitation of the RE-FWM process is the available power at 202.38 nm. The UV light is difficult to generate and there are several transmission losses in the optics. Furthermore, the requirement of collinearity of the two beams is a limitation since chromatic aberration impedes the realization of the optimal configuration.

The THG system being implemented uses a diode cw laser at 729 nm.
5.1. Lyman-α Source

This laser is amplified in a Ti:Sapphire pulse amplifier pumped by an Nd:YAG pulsed laser. The generated 729 nm is further doubled in a SHG crystal and tripled in a Kr:Ar mixing chamber. Fig. 5.1 shows an schematic of the system. Before delivery into the ALPHA trap the 364 nm will be filtered with a narrow band Lyman-α filter. This system is being implemented and is expected to be completed during the fall of 2014. The expected conversion efficiency is $\sim 10^{-5}$. Optical cooling experiments are expected to start in the summer of 2015.
5.1. Lyman-α Source

Figure 5.1: Since experiments at ALPHA require a very stable and robust system it was decided that the THG process would be a better candidate. The input laser required for the THG can be implemented with solid state lasers. This type of system is more robust and requires less maintenance that a dye laser based system. The THG system being implemented uses a diode cw laser at 729 nm. This laser is amplified in a Ti:Sapphire pulse amplifier pumped by an Nd:YAG pulsed laser. The generated 729 nm is further doubled in a SHG crystal and tripled in a Kr:Ar mixing chamber.
5.2 Alternative hydrogen source

By pulsing a high voltage across a pair of electrodes positioned in front of a valve during supersonic expansion it is possible to create a temporary discharge plasma. If the electron density and electron energy is sufficiently high, a large number of the expanding molecules can be dissociated. The technique is usually implemented with an external filament. This facilitates the plasma creation by supplying a large number of free electrons. We have used this technique to create hydrogen by dissociating pure and seeded CH$_4$ and H$_2$. As a seed gas we have used Ar and Kr. We have verified the dissociation of hydrogen with the QMS. Unfortunately, this discharge technique has been found to deplete the beam and reduce the total density of atoms in it. A possible explanation is that the plasma generated is diverged by the fringing electric field around the electrodes. Fig. 5.2 shows the ratio of the signal intensity for various masses as a function of the discharge voltage applied. Another problem with plasma expansion is that the discharge process heats the gas increasing the transversal speed and the longitudinal spread of speed.

Theoretically, it is possible to optimize the discharge so the beam is not depleted. As an example we can look at dissociation of pure molecular hydrogen. There are a few processes that can produce atomic hydrogen in plasma. In principle molecular dissociation has to compete with an ionization process. If the distribution of electron energies is wide, the rate at which any reaction proceeds depends mostly on the onset energy and the cross section immediately beyond onset. If the distribution is very narrow then the maximum cross section can be used to increase the probability of a specific reaction $[137]$. The following table shows some of the main hydrogen dissociation reactions ($[137]$):

In the table reaction 2 and 3 can be neglected because of too high onset energy. The atomic hydrogen is mainly produced by the reactions 4, 5 and 6 and since the density of molecular ions is much smaller than that of molecules the dominant process is reaction number 5. Also the other processes are not very probable because there are not enough excited re-
5.2. Alternative hydrogen source

Figure 5.2: Discharge plates have been used to generate hydrogen by dissociating pure and seeded CH$_4$ and H$_2$. As a seed gas we have used Ar and Kr. We have verified the dissociation of hydrogen with the QMS. Unfortunately, this discharge technique has been found to deplete the beam and reduce the total density of atoms in it. A possible explanation is that the plasma generated is diverged by the fringing electric field around the electrodes.
5.2. Alternative hydrogen source

Table 5.1: Onset energy of electrons and maximum cross section for hydrogen dissociation processes in the literature.

<table>
<thead>
<tr>
<th>No</th>
<th>Process</th>
<th>Onset (eV) (eV)</th>
<th>$\sigma_{\text{max}} \times 10^{-16}$ (cm$^2$)</th>
<th>Energy at $\sigma_{\text{max}}$ (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\text{H}_2 + e \rightarrow \text{H}_2^+ + 2e$</td>
<td>15.4</td>
<td>1.1</td>
<td>80</td>
</tr>
<tr>
<td>2</td>
<td>$\text{H}_2 + e \rightarrow \text{H}^+ + \text{H} + 2e$</td>
<td>18.0</td>
<td>0.005</td>
<td>120</td>
</tr>
<tr>
<td>3</td>
<td>$\text{H}_2 + e \rightarrow \text{H}^+ + \text{H}^+ + 3e$</td>
<td>46</td>
<td>0.005</td>
<td>120</td>
</tr>
<tr>
<td>4</td>
<td>$\text{H}_2^+ + e \rightarrow \text{H}^+ + \text{H} + e$</td>
<td>12.4</td>
<td>3-16</td>
<td>16</td>
</tr>
<tr>
<td>5</td>
<td>$\text{H}_2 + e \rightarrow \text{H} + \text{H} + e$</td>
<td>$\approx$ 8.5</td>
<td>0.6</td>
<td>12</td>
</tr>
<tr>
<td>6</td>
<td>$\text{H}_2^+ + e \rightarrow \text{H} + \text{H}$</td>
<td>0</td>
<td>100(?)</td>
<td>–</td>
</tr>
<tr>
<td>7</td>
<td>$\text{H} + e \rightarrow \text{H}^+ + 2e$</td>
<td>13.5</td>
<td>0.65</td>
<td>40</td>
</tr>
<tr>
<td>8</td>
<td>$\text{H} + e \rightarrow \text{H}^+(2p) + e$</td>
<td>10.2</td>
<td>0.7</td>
<td>25</td>
</tr>
<tr>
<td>9</td>
<td>$\text{H}^+ + e \rightarrow \text{H}^+ + 2e$</td>
<td>3.3</td>
<td>15</td>
<td>9</td>
</tr>
<tr>
<td>10</td>
<td>$\text{H}_2 + e \rightarrow \text{H}_2^+ + e$</td>
<td>10.3</td>
<td>0.2</td>
<td>60</td>
</tr>
</tbody>
</table>

action partners. Given that the cross section of reaction 5 is quite small a large density of energetic electrons is necessary to produce a sizeable yield of hydrogen. If the voltage applied is reduced, and a large amount of monochromatic electrons is supplied to the beam, it is possible to increase the rate of a specific reaction without depleting the beam. This is possible by replacing the thermionic electron emitter (filament) by a cold field electron emitter. By selecting a material with a low work function and a suitable geometry it is possible to extract electrons via quantum tunneling (monochromatic electrons) with low electric field. The best material available for such application is carbon nanotubes [138, 139]. Carbon nanotubes are allotropes of carbon with a tubular structure. The carbon atoms are arranged in a honeycomb lattice that can be thought of as rolled graphene. Nanotubes can have a very high aspect ratio. For instance, nanotubes synthesized with length-to-diameter ratios of up to 28,000,000:1 have been reported [140]. Single-walled nanotubes (SWNTs) are formed by one sheet of graphene. Multi-walled nanotubes (MWNTs) are formed by
5.2. Alternative hydrogen source

several SWNTs in a coaxial arrangement (several concentric SWNTs form one MWNT). Nanotubes are formed entirely from $sp^2$ hybrid bonds as in graphene. This bonding structure is stronger than the $sp^3$ bonds found in diamond and that makes them a particularly strong and stable material. These properties combined with the high conductivity found in nanotubes make them a very good candidate for cold field-emitter sources. Due to their high aspect ratio, an externally applied electric field is enhanced hundreds to thousands of times at their tip. It is thus possible to extract electrons with low applied voltages and, since the nanotube tip radius is on the order of a few nanometers, it is possible to produce very localized electron sources. Low-voltage operation combined with the monochromaticity of the electrons produced by quantum tunnelling makes them the perfect candidate for our application. Fig. 5.3 shows two carbon nanotube based discharge valves implemented as a preliminary test of this technique. In the first implementation an array of MWNTs (called a forest) on a silicon substrate was used as the cathode of two electrodes placed across a supersonically expanding beam. In the second implementation a stainless steel mesh is used as anode and two carbon nanotube forests are used as the cathode. The principle of operation is that some of the electrons emitted would accelerate past the mesh and dissociate molecules expanding by collision. Since the expanding gas is in a region of no potential gradient it should not experience any depletion. In order to implement the discharge sources the carbon nanotube forests had to be fabricated. This was done with a chemical vapour deposition process and the development of this project with examples of the nanotubes grown in stainless steel and silicon substrates are discussed in Appendix B.
5.2. Alternative hydrogen source

Figure 5.3: Carbon nanotube based discharge valve prototypes. The first prototype (top) used an array of MWNTs (called a forest) on a silicon substrate as the cathode of two electrodes placed across a supersonically expanding beam. In the second implementation a stainless steel mesh is used as the anode and two carbon nanotube forests are used as the cathode. The principle of operation is that some of the electrons emitted would accelerate past the mesh and dissociate molecules expanding by collision. Since the expanding gas is in a region of no potential gradient it should not experience any depletion.
5.3 Conclusion

The main topic of this dissertation was the implementation of a Lyman-α source based on a RE-FWM process. For this work a particular strong resonance on krypton \((4s^2 4p^5 5p[1/2]_o \leftarrow 4s^2 4p^6(1S_0))\) was used. The optimization of the source was performed by changing the phase matching parameters such as the gas mixture, used to accomplish the condition \(\Delta k = 0\), and the focusing geometry. In addition, the light produced by parasitic processes was monitored. The power of the source was measured indirectly using the LIF of hydrogen. The experimental powers measured are summarized in Table 5.2. These power levels are sufficient for optical cooling of antihydrogen which is the motivation for this thesis project as described in Chapter 1. The linewidth of the source was estimated to be 6.6 GHz. While the power is sufficient for the described objectives, the source linewidth needs to be narrowed for optical cooling.

<table>
<thead>
<tr>
<th>Energy ((\mu J/pulse))</th>
<th>Repetition ((Hz))</th>
<th>Pulse Length ((nsec))</th>
<th>Power ((\mu W))</th>
<th>Linewidth ((GHz))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.125 ± 0.030</td>
<td>10</td>
<td>9</td>
<td>1.25 ± 0.30</td>
<td>6.6</td>
</tr>
<tr>
<td>0.025 ± 0.006</td>
<td>50</td>
<td>9</td>
<td>1.25 ± 0.30</td>
<td>6.6</td>
</tr>
</tbody>
</table>

Along with the RE-FWM implementation a THG source was also implemented. To measure the power of the THG source an acetone ionization detector was implemented. The maximum power generated was \(\sim 0.6 \mu W\). However, the input laser can easily be doubled and even tripled in power. Also it is possible to implement a solid state system with this process. This is favourable with regards to stability and maintenance in contrast with a dye laser system. These results show that THG can potentially be used to generate Lyman-α for cooling of antihydrogen. In addition, the implementation of two lasers, a RE-FWM and a THG, has provided a tangible
5.3. Conclusion

comparison of the potential of each technique. Based on the implementation experience and results, it was decided that THG is a more feasible system for the antihydrogen cooling.

Chapter 4 described the implementation of a hydrogen beam. The hydrogen beam was implemented by entrainment of hydrogen, produced with a gas cracker, in a supersonic expansion of Ar. The hydrogen was confirmed with a QMS. A Zeeman bender was used to separate the hydrogen from the combined beam (Ar, H$_2$ and H) as part of the confirmation. Doppler free LIF spectroscopy of the hydrogen beam was attempted and some signatures of fluorescence were obtained. In addition, the supersonic beam was used to demonstrate the potential of a MOT as a very localized pressure (density) measuring device. The MOT atom-loss-rate during a collision with the beam provides information of the beam density and velocity distribution. By measuring the scattered light from the MOT it is possible to quantify the loss-rate, and in that manner to measure the beam density. Experiments with Ar, Kr and He were used to demonstrate the measuring technique.
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Appendix A

Characterization of solenoids in the ALPHA trap

In the introduction it was mentioned that the ALPHA trap has laser access only along one dimension. To perform 3D cooling with one laser beam it is necessary to couple the 3 degrees of freedom. The dynamical coupling between the $z$ and $xy$ degrees of freedom can be achieved by the use of non-harmonic magnetic fields. The nonlinearity is produced along the $z$ direction by using five solenoidal coils. The ALPHA trap uses two mirror coils to provide axial confinement and an octupole to create radial confinement. The antihydrogen in an LFS state is trapped at the centre in the magnetic minimum. The five solenoids are placed between the mirror coils. The 3 solenoids at the centre provide a flatter magnetic field instead of the quadratic dependence that it would have otherwise. The effective potential has a $\sim r^6$ potential along the $z$ axis and in the radial direction in this trap.

During repeated trapping of antihydrogen the coils have to be quenched so it is necessary to understand the coupling between the coils to obtain the desired magnetic field. In this appendix the characterization of the coupling between the solenoids is discussed. Using PSIM, a SPICE software, the solenoids and mirrors coils were simulated during quenching. The software solves the differential equations of the RCL circuits taking into account the couplings between the coils (mutual inductance). The circuit is modelled with measured values for all the different components. To verify how well the model represents the real device some experimental measurements were recreated. The mirror coils are refereed as MA, MB, MC and MD; the solenoids as S1 and S2. Given the symmetry and the poor
Appendix A. Characterization of solenoids in the ALPHA trap

coupling between third nearest neighbours coils (for instance MA and MD) it is possible to accurately represent the real circuit by simulating only half of the coil array. The simulation includes S1, MA, MB and MC with its respective RCL circuits. The inductive parts of MD and ME have also been included. However, removing the MD and ME coils does not change the results. Tables A.1, A.2 shows the values that were used in the circuit simulation:

Table A.1: ALPHA coils inductances and mutual inductances.

<table>
<thead>
<tr>
<th>(mH)</th>
<th>S1</th>
<th>MA</th>
<th>MB</th>
<th>MC</th>
<th>MD</th>
<th>ME</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>24.17</td>
<td>0.457</td>
<td>0.0075</td>
<td>0.005</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>MA</td>
<td>1.032</td>
<td>0.075</td>
<td>0.00015</td>
<td>0.0007</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>MB</td>
<td>1.057</td>
<td>0.078</td>
<td>0.001</td>
<td>0.0005</td>
<td></td>
<td></td>
</tr>
<tr>
<td>MC</td>
<td>1.054</td>
<td>0.079</td>
<td>0.001</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MD</td>
<td>1.055</td>
<td>0.072</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ME</td>
<td>1.035</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table A.2: ALPHA coils parasitic capacitance and series resistance.

<table>
<thead>
<tr>
<th>Coil</th>
<th>$C_p$ (µF)</th>
<th>$R_s$ (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>1.116</td>
<td>15.4</td>
</tr>
<tr>
<td>MA</td>
<td>21.23</td>
<td>1.9</td>
</tr>
<tr>
<td>MB</td>
<td>21.22</td>
<td>2.1</td>
</tr>
<tr>
<td>MC</td>
<td>21.26</td>
<td>2.0</td>
</tr>
</tbody>
</table>

The source in the simulation was a 5 $V_{pp}$, 50 Hz sinusoidal. The voltage induced was measured in the adjacent coils across a 1 MOhm resistor. In the real experiment when the source was applied to MA a voltage of 360 mV ($V_{pp}$) and 59.2 mV ($V_{pp}$) were measured across S1 and MB respectively.
Appendix A. Characterization of solenoids in the ALPHA trap

Figure A.1: ALPHA coils parameter measurement simulation. The source in the simulation was a $5\, \text{V}_{pp}$, 50 Hz sinusoidal. The voltage induced was measured in the adjacent coils across a 1 MOhm resistor.
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The simulation reproduces the experiment very accurately ($V_{S1} = 370$ mV and $V_{MB} = 61.2$ mV in the Fig. A.1 and Fig. A.2).

Figure A.2: In the real experiment when a 5 $V_{pp}$, 50 Hz sinusoidal source was applied to MA a voltage of 360 mV ($V_{pp}$) and 59.2 mV ($V_{pp}$) were measured across S1 and MB respectively.

Resistors in parallel can be used as intrinsic protection to reduce the current induced in the mirror coils during a quench. The voltage induced across the resistor should also be reduced as to avoid arching (and possible damage of the insulation). To induced a quench it usually requires some active switch and a dissipative element. The dissipative quenching resistor ($R_Q = 10$ Ohm) was chosen so the decay time was $\sim 10$ msec. The quench was initiated by opening the IGBT in the driving circuit. In this simulation a small series resistance of 1 mOhm was added to the circuit to account for
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the lead resistance (see Fig. [A.3]).

Fig. [A.4] shows the IGBT gate driving voltage (V1), the current decay during the quench in S1 ($I_{S1}$), and the induced current in MA ($I_{QA}$) and MB ($I_{QB}$) with identical protection resistors $R_p = 1$ ohm and with an initial current $I_{S1} = 250$ Amps.
Figure A.3: The dissipative quenching resistor ($R_Q = 10$ Ohm) was chosen so the decay time was $\sim 10$ msec. The quench was initiated by opening the IGBT in the driving circuit. In this simulation a small series resistance of 1 mOhm was added to the circuit to account for the lead resistance.
Figure A.4: IGBT gate driving voltage ($V_1$), the current decay during the quench in $S_1$ ($I_{S1}$), and the induced current in $A$ ($I_{QA}$) and $B$ ($I_{QB}$) with identical protection resistors $R_p = 1$ ohm and with an initial current $I_{S1} = 250$ Amps.
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During the experiment it is more likely that a quench of Mirror coils A or E is induced. The quench of MA was simulated using a quench resistor ($R_pA = 0.5 \text{ Ohm}$) that induces a decay time of 10 msec. The induced current in S1 and MB were simulated using protection resistors ($R_pS1$ and $R_pB$) of 10 Ohms (see Fig. A.5). The capacitance, as before, did not affect the circuit at this decay time.

According to the simulation (Fig. A.6) a 10 msec quench in MA or MD starting from an initial current of 800 Amps would induce a peak current of 6 Amps in S1 and 0.33 Amps in MB (with respective voltages of 60 V and 3.3 V) when 10 Ohm protection resistors are used. The induced current decays at the same rate as the quenching current in about 10 msec. This is convenient in S1 as the same protection resistor could be the quenching resistor (quench of 10 msec in S1, $R = 10 \text{ Ohm}$) providing an intrinsic protection (without requiring active elements).
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Figure A.5: The quench of MA was simulated using a quench resistor ($R_{pA} = 0.5$ Ohm) that induces a decay time of 10 msec. The induced current in S1 and MB were simulated using protection resistors ($R_{pS1}$ and $R_B$) of 10 Ohms.
Figure A.6: A 10 msec quench in MA or MD starting from an initial current of 800 Amps would induce a peak current of 6 Amps in S1 and 0.33 Amps in MB (with respective voltages of 60 V and 3.3 V) when 10 Ohm protection resistors are used. The induced current decays at the same rate as the quenching current in about 10 msec.
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If a quench were required in S1, 10 msec decay from 250 Amp would induce 25 Amps in MA and 1.2 Amps in MB (with respective voltages of 25 V and 1.2 V) when a 1 Ohm protection resistor is used. The quench resistor that causes a 10 msec decay in MA is 0.5 Ohm. If intrinsic protection of MA is required while having the ability to quench S1 and MA with 10 msec decay a possible arrangement is the following:

- Resistors in parallel in S1 and S2 with total R = 10 Ohm and P = 1000 W for quenching and protection from induced current during a MA or ME quench.

- Resistors in parallel in MA and ME with total R = 0.5 Ohm and at least P = 1000 W for quenching and protection from induced currents during a S1 or S2 quench.

- Resistor in parallel in MB and MC with total R = 10 Ohm and P = 10 W.

Fig. A.7 is a simulation of that scenario while quenching S1 (the same configuration while quenching MA can be seen in Fig. A.6).
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Figure A.7: A 10 msec quench in S1 starting from an initial current of 250 Amps. Resistors in parallel in S1 with total $R = 10$ Ohm and $P = 1000$ W for quenching and protection from induced current during a MA or ME quench. Resistors in parallel in MA with total $R = 0.5$ Ohm and at least $P = 1000$ W for quenching and protection from induced currents during a S1 quench.
Appendix B

Fabrication of carbon nanotube electron sources

In order to implement the electron sources mentioned in Chapter 5 arrays of MWNT’s have been synthesized (grown) on silicon and stainless steel substrates. The ideal array of nanotubes consists of multiple, vertical and parallel nanotubes. Chemical Vapour Deposition (CVD) is the most common method used for growing carbon nanotubes. The CVD technique requires nanoparticles of a metal catalyst, usually iron, nickel or cobalt, that provides a base for the nucleation of the nanotubes. The carbon is provided in the form of a carbon containing precursor gas such as acetylene, ethylene or methane. To grow nanotubes on silicon substrates a thin layer of the metal catalyst (∼1 nm) is deposited using e-beam evaporation. In this case, iron was used as the catalyst. The silicon substrates were placed in the middle stage of a three stage furnace. At the reaction temperatures, usually 600-800°C, the carbon diffuses on the surface of the metal nanoparticles and forms the carbon nanotube rings. Growth on silicon is very reliable and Fig. B.1 shows scanning electron microscope (SEM) and transmission electron microscope (TEM) pictures of the samples grown in silicon. The nanotubes grown this way are approximately 2 - 4 mm long. We have used these nanotubes for the prototype discharge valve shown in Chapter 5. However, the electrical contact between the nanotube and the silicon substrate is not very good since the silicon has a very thin layer of SiO on the surface which is an insulator.

In order to improve the electrical contact nanotubes were grown on stainless steel. Polished sheets of stainless steel 304 were sonicated and
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etched in 37% HCl (Sigma-Aldrich) prior to CVD. It is possible that the acid exposes the iron grains. With the heat treatment some of the iron form nanoparticles from which the nanotubes grow. Fig. B.2 shows SEM and TEM pictures of nanotubes grown on stainless steel. These nanotubes do not form vertical arrays. It is believed this is caused by the surface roughness of the substrate.

Finally, we are currently attempting to grow nanotubes on nickel alloys and to improve the surface of the stainless steel by electropolishing during the etching. All the nanotube growth was performed in Dr. Takamasa Momose lab by Bill Wong under my supervision.
Figure B.1: SEM and TEM pictures of carbon nanotube array grown on silicon substrates. The TEM picture indicates MWNT’s with several concentric nanotubes.
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Figure B.2: SEM and TEM pictures of carbon nanotube array grown on stainless steel substrates. The TEM picture indicates MWNT’s with a bamboo-like structure.