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RYDBERG EXCITATION OF HYDROGEN FOR STUDIES TOWARDS ANTIHYDROGEN DEEXCITATION

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STATEMENT OF AUTHORSHIP

I hereby certify that this bachelor thesis has been composed by myself and describes my own work unless otherwise acknowledged in the text. All references and verbatim extracts have been quoted, and all sources of information have been specifically acknowledged. It has not been accepted in any previous application for a degree.

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ABSTRACT

The CERN Antiproton Decelerator (AD) facility hosts antimatter experiments, which produce antihydrogen in a wide range of Rydberg states. Precision measurements require groundstate antiatoms that are currently only available through spontaneous decay. The antihydrogen experiments can be categorised as trap and beam experiments. The trap experiments use a magnetic field to accumulate atoms and let them decay before performing measurements, but the beam experiments like AEGIS and ASACUSA cannot afford to wait for the antiatoms to spontaneously decay.

The group I joined for my bachelor thesis is investigating methods to deexcite highly excited antiatoms within a few tens of μs . Several deexcitation schemes have been proposed. However, before they are employed on antihydrogen, a proof-of-principle experiment on hydrogen has been developed. It aims to produce Hydrogen atoms in highly excited states before demonstrating its deexcitation.

In this work, I was involved in both experimental and theoretical aspects. On the experimental side, I helped set up the beamline and the Doppler-spectroscopy diode and Ti:Sa excitation laser, wrote a data-acquisition program, took spectroscopic data of the hydrogen source, and analysed them. The spectroscopy data helped the characterisation of the plasma. On the theoretical side, I used a code to find the optimal E and B fields and their mutual orientations to produce circular states via the cross-field methods. The results will guide the experimental implementation of the method.

Dedicated to all my teachers, and parents.

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I will eternally be grateful to my 'Gurus': **Prof. Dr. Johannes Deiglmayr** and **Dr. Chloé Malbrunot**, for they are the teachers I always yearned. I need to say that I couldn't have gotten a more diligent mentor at the university than Prof. Deiglmayr. His responses were always insightful and quick. I will always look up to him as a great researcher and teacher. I am thankful to Dr. Malbrunot for her patient involvement in solving innumerable problems that I faced (for more than a year) before finally making it to CERN. I am grateful to her for arranging funds for my extended stay.

I feel fortunate for having mentors who supervised me meticulously. Throughout the last five months, I learned a lot from both of them, not just about physics but also about a good researcher's traits. With that said, I will try my best to manifest all the qualities of my mentors. I hope, one day, to be the student who makes them proud.

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*"The people who are crazy
enough to think that they can change
the world, are the ones who do."*

-Steve Jobs

"Arise, awake, stop not until the goal is reached."

-Swami Vivekananda

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Chapter One

Introduction

1.1 The Antimatter Factory at CERN and Overview of the Physics Program

CERN hosts a so-called "Antimatter Factory" (ref. fig. 1.1) that includes the [Antiproton Decelerator](#) (AD) and [ELENA](#) (Extra Low ENergy Antiproton) (ref. fig. 1.1). AD and ELENA are unique machines that are capable of producing low-energy antiprotons. The AD can produce antiproton of 5.3 MeV. ELENA is a new deceleration ring integrated with the AD that can slowdown the antiprotons further to 0.1 MeV. It has a circumference of 30 m.

Antiprotons are required to synthesise antihydrogen atoms, the simplest atoms made entirely of antimatter which is one of the main research focus at the AD. Antiprotons are produced as one of the secondary particles when energetic protons from the PS are fired onto a metal block. Before antiprotons can be used to formed antiatoms, they need to be slowed down and trapped. These antiprotons can then be utilised by the antimatter experiments in the AD hall either by directly performing measurements on the antiprotons or by synthesising antiatoms. So far, only antihydrogen has been synthesised.

Currently, the antimatter factory hosts five experiments: $\text{AE}\bar{g}\text{IS}$, ASACUSA, ALPHA, BASE and GBAR.

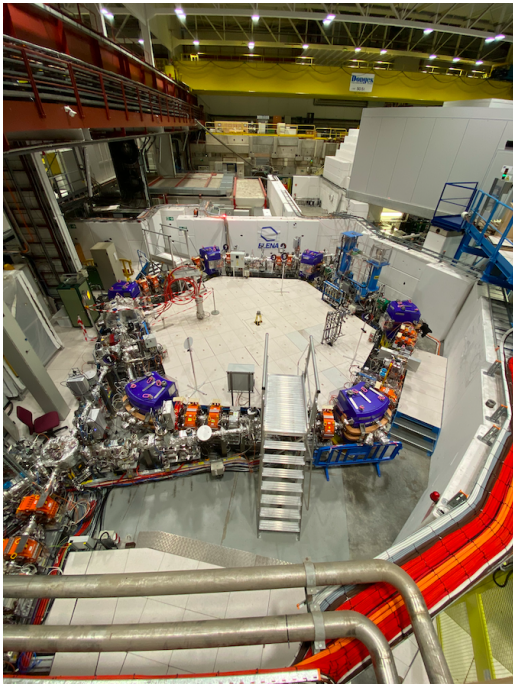
1. $\text{AE}\bar{g}\text{IS}$ aims to study interaction of gravity and antimatter by testing the Weak Equivalence Principle(WEP).
2. ASACUSA does spectroscopy on antihydrogen and antiprotonic helium to test CPT symmetry.
3. ALPHA traps antihydrogen atoms and performs spectroscopic studies. It also aims at testing antimatter gravity with trapped antihydrogen.
4. BASE aims to measure the (anti)proton magnetic moment and charge to mass ratio to look for differences between matter and antimatter.
5. GBAR intends to test the Weak Equivalence Principle(WEP) with antihydrogen just like $\text{AE}\bar{g}\text{IS}$ but antihydrogen is produced at a few tens of μK by cooling $\bar{H}^+$ ions.

This thesis will discuss details relevant to the $\text{AE}\bar{g}\text{IS}$ and ASACUSA experiments.

1.2 $\bar{H}$ Beam Experiments: $\text{AE}\bar{g}\text{IS}$ and ASACUSA

Before discussing the $\text{AE}\bar{g}\text{IS}$ and ASACUSA experiments in depth, first I discuss their main goals and the theories they aim to test.

The aim of the $\text{AE}\bar{g}\text{IS}$ (Antihydrogen Experiment: Gravity, Interferometry, Spectroscopy) experiment is to understand the interaction of gravity with antimatter to test the WEP (Weak Equivalence Principle). The WEP is one of the founding principles of General Relativity and it predicts that all particles should have the same inertial and gravitational



(a) The ELENA decelerator



(b) The Antimatter factory at CERN

Figure 1.1: (a) The ELENA(Extra Low ENergy Antiproton) decelerator in the Antimatter factory at CERN is capable of producing very low energy antiproton beams that can be trapped for antihydrogen production. (b) The Antimatter factory at CERN hosts five antimatter experiments currently: $AE\bar{g}IS$, ASACUSA, BASE, ALPHA and GBAR.

mass. The WEP has been tested exclusively on matter systems with an accuracy of 10^{-14} and it is yet to be experimentally tested on antimatter¹.

The ASACUSA (Atomic Spectroscopy And Collisions Using Slow Antiprotons) experiment aims at testing CPT symmetry by probing antihydrogen ground-state hyperfine structure, and by determining the antiproton to electron mass ratio in antiprotonic Helium. According to the CPT (Charge conjugation, Parity, Time-reversal) theorem², any Lorentz invariant local quantum field theory with a Hermitian Hamiltonian should conserve CPT symmetry. Some consequences of this theorem are that the properties of matter and antimatter, like mass, lifetimes etc should be the same, while electrical charges should be opposite. To test the CPT theorem, properties of matter and antimatter systems need to be precisely measured. CPT tests are performed on both atomic and sub-atomic systems e.g., the BASE experiment at CERN that studies antiprotons. Atomic systems have the advantage that their energy transitions can be measured with relatively high precision e.g., the 1s-2s transition of magnetically trapped $\bar{H}$ has been compared with the theoretical value of the same transition in H with an accuracy of 2×10^{-12} .³

Both AE $\bar{g}$ IS and ASACUSA experiments do not trap antihydrogen. Instead, they use antihydrogen beam for performing measurements. Trap and beam experiments mainly differ in the following ways: Beam experiments can work with atoms of relatively high temperatures (state of the art in antihydrogen formation temperature is currently ~ 50 K), unlike trap experiments (e.g. ALPHA) that can only trap atoms at ~ 0.5 K and lower. Measurements of hyperfine transitions are more prone to errors in trap experiments due to the presence of inhomogeneities in the trapping magnetic field. The solid angle of the measurement region is smaller compared to the antihydrogen formation region. Thus, to perform measurements, a focused beam of atoms has to be formed. Without a focused

beam, the antiatoms will annihilate on the beamline walls. For this purpose, ASACUSA uses magnetic focusing; AE $\bar{g}$ IS envisions stark acceleration. The trap experiments can hold atoms until they spontaneously decay, but the beam experiments cannot.

Antihydrogen atoms used in AE $\bar{g}$ IS and ASACUSA experiments are produced through the following two mechanisms:

1. **Three-Body-Recombination (3BR):** In this mechanism two positrons and an anti-proton react to form antihydrogen. The second positron carries away excess energy of formation.



At the AD, the antiprotons and positrons are simultaneously trapped in a Penning trap and are brought together using different techniques. Usually those techniques lead to a so-called "continuous" production of antihydrogen, meaning that the production time of $\bar{H}$ is not well known within the typical duration of interaction between the antiprotons and positrons (in the range of several hundreds of milliseconds).

3BR produces Rydberg antihydrogen in a broad range of n (theoretically up to $n=100$). ASACUSA uses 3BR for antihydrogen production.

2. **Charge-Exchange (CE):** This mechanism involves positronium (Ps: a bound state formed by an electron and a positron) that releases positive charge to the trapped and cooled antiproton. This reaction produces antihydrogen and the remaining electron carries away excess energy.



In the AE $\bar{g}$ IS experiment, the CE process is used with previously laser-excited Ps atoms

¹* denotes Rydberg state

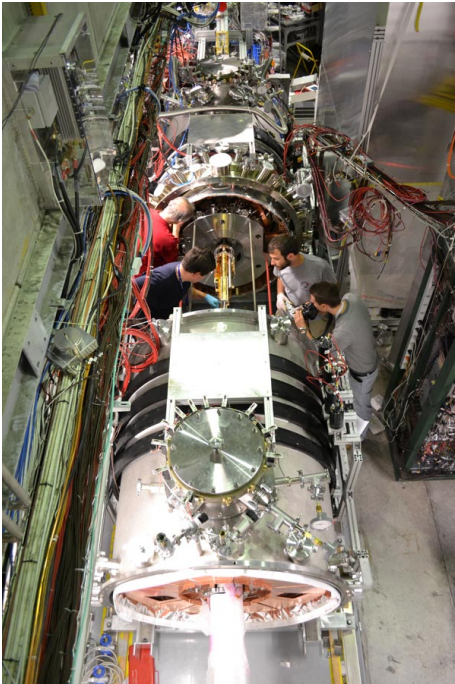
(using a UV and infrared laser pulse). The excitation step of Ps^* adds complexity to the process but it dramatically increases the cross-section (proportional to n^4). Without excitation there would be very little antihydrogen production. The other advantage is that a narrower range of antihydrogen n states can be targeted since, via the CE mechanism, Ps "imprints" its quantum state onto the formed antihydrogen atoms. Finally, CE allows time tagging of antihydrogen production determined by the Ps excitation laser timing. This is a key point for AEGIS, which relies on precise timing to determine how antihydrogen behaves in the Earth's gravitational field. So, antihydrogen production through CE is rather a necessity.

Recently in AE $\bar{g}$ IS, the CE reaction was implemented to produce pulsed antihydrogen. The time when 90% of the atoms were produced was measured with an uncertainty of ~ 250 ns⁴.

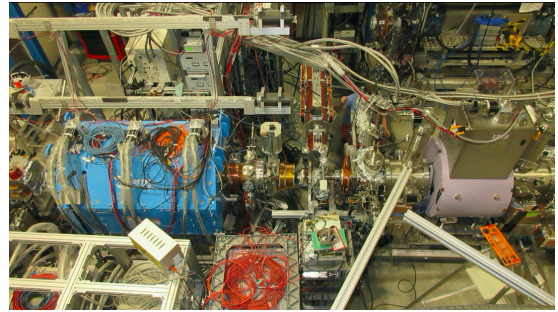
Both 3BR and CE processes form Rydberg antihydrogen distributed overall l, m values with spontaneous lifetimes given by Eq. 1.3.⁵ With this equation, the highest antihydrogen lifetimes can be approximated to tens of milliseconds. These correspond to the highest l states near $n \sim 30$. The highest l states in n manifolds are called the circular states (ref. section 2.1.1) and they have the highest lifetimes due to n^5 proportionality compared to n^3 for low l states.

GS atoms are required to perform measurements and antihydrogen atoms, after formation, have velocities of ~ 1000 m/s^{6,7}. If not deexcited within tens of μ s, then no GS antiatoms would be available in the measurement region. That is why, stimulated deexcitation techniques for $\bar{H}$ have been proposed like in ref.⁶.

$$\tau \approx \left(\frac{n}{30}\right)^3 \left(\frac{l+1/2}{30}\right)^2 \times 2.4 \text{ ms} \quad (1.3)$$



(a) The AE $\bar{g}$ IS experiment²



(b) The ASACUSA-CUSP apparatus³

Figure 1.2: The AE $\bar{g}$ IS experiment studies interaction of gravity with matter to test the WEP. The ASACUSA experiment has two focuses: CPT symmetry test on Antiprotonic helium and Antihydrogen.

1.3 Stimulated Deexcitation of $\bar{H}$

Theoretical studies on ways to rapidly stimulate the deexcitation of antihydrogen atoms have been performed. One way is through electric and magnetic field mixing of the Rydberg population⁸. States belonging to $n = 20 - 30$ manifolds in antihydrogen were fully mixed using electric and magnetic fields. Then, a laser was used to deexcite the occupied states to $n = 3$ after which they spontaneously decayed to the ground-state. It was found that $\sim 60\%$ of antihydrogen atoms can be deexcited to ground state within $10\,\mu\text{s}$ using a broadband 5000 GHz laser producing $\sim 10\,\text{J}$ ($I = 1.8 \times 10^{10}\,\text{W/m}^2$) at 828 nm. It was found that fields of intensities $3n|F| \sim |B|$ ^{8,9} were optimal to mix states. Other deexcitation techniques have been proposed using a combination of THz, microwave and optical light⁶.

1.4 Hydrogen Experimental Setup for Proof-of-Principle Experiment of Stimulated Deexcitation

Before applying deexcitation techniques to antihydrogen, a proof of principle experiment on hydrogen has been developed. In order to demonstrate fast stimulated deexcitation of Rydberg hydrogen, a beam of excited atoms has to be produced and that is essentially the aim of the H excitation experiment.

The experimental setup consists of a hydrogen discharge plasma source producing a large amount of ground state hydrogen atoms in an Evenson microwave cavity. Rydberg states have been detected downstream of the source by field ionization techniques indicating that some higher n-states are produced and make it out of the plasma. In principle, this

²Adapted from <https://aegis.web.cern.ch/aegisMultimedia.html>

³Adapted from <https://asacusa.web.cern.ch>

observation can indicate an even larger population of the 2S state. If confirmed, that would enable Rydberg excitation directly from the 2S state to $n = 30$ via a ~ 366 nm laser. If the amount of 2S state in the beam is found to be too small, a number of other techniques could also allow the excitation from GS to 2S or 2P: 2-photon excitation, Lyman-alpha excitation, population of the 3P using a lamp that decays to 2S etc.

For the excitation to Rydberg state, $n=30$ was chosen as a target for several reasons related to the deexcitation mechanisms using THz light (ref.⁶). The laser setup to generate the ~ 366 nm light consists of a Nd:YAG pump laser that produces 532 nm pulsed laser beam which passes through a Ti:Sapphire gain cavity and is converted to ~ 732 nm beam. It is seeded with a diode laser. At last, it is frequency doubled with a BBO (Beta barium borate) crystal to ~ 366 nm. This beam is fed perpendicularly into a beamline where it excites atomic H. After exciting H atoms, they fly towards a detection chamber where two metallic grids of opposite charge polarity connected to a high-voltage power supply rip the bounded electrons and protons. Their count is detected by two Microchannel Plates (MCPs) located perpendicularly to the hydrogen beam path (ref. Chapter 4).

Excitation of electron into the circular state (highest l, m) is not possible with only a laser because dipole selection rules do not allow transitions other than $\Delta l = \pm 1$ so a 2s state can only be excited to 30p state using a laser. Introducing external electric ($\mathbf{F}$) and magnetic ($\mathbf{B}$) field with a certain mutual orientation can induce maximum mixing of states. In other words, the degeneracy of energy levels vanishes and selection rules change. In this situation, l does not remain a good quantum number and the parabolic quantum numbers n, k, m come into play. A simulation has been done (ref. Chapter 3) in which $\mathbf{F}$ and $\mathbf{B}$ field intensities were varied, along with their mutual orientation to maximise transition rates to the circular state.

Chapter Two

Theoretical Background

2.1 Rydberg Atoms

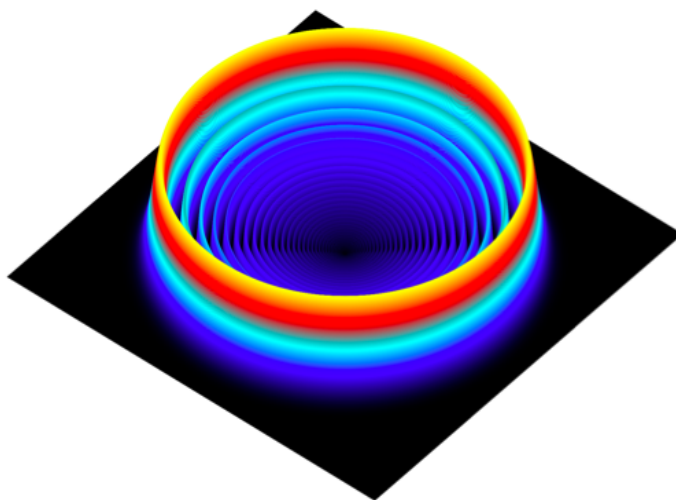


Figure 2.1: Probability density distribution of 35s Rydberg hydrogen¹⁰

Rydberg atoms are atoms in highly excited states, that is in states with high principal quantum number n . They exhibit many exaggerated properties like large polarisabilities, dipole moments, and long radiative lifetimes. These exaggerated properties are the consequence

of the huge extent of the electronic wavefunctions of Rydberg states. The energy levels of hydrogen are given by $E_{nl} = -\frac{E_h}{2n^2}$ where E_h is the binding energy of hydrogen in ground state (-13.6eV). This value is also known as the Rydberg constant.

Some of the properties of Rydberg atoms have been summarised in the table 2.1.

n Dependence of Some Properties
Energy Spacing $\propto \mathbf{n}^{-3}$
Binding Energy $\propto \mathbf{n}^{-2}$
Dipole moment $\propto \mathbf{n}^2$
Orbital radius $\propto \mathbf{n}^2$
Radiative lifetime $\propto \mathbf{n}^3$
Cross-section $\propto \mathbf{n}^4$
Polarizability $\propto \mathbf{n}^7$

Table 2.1: Dependence of some properties of Rydberg atoms¹¹ on $\mathbf{n}$. $\mathbf{n}=n-\delta_{nl}$ is the effective principal quantum number, where δ_{nl} is the empirical quantum defect. Detailed description can be found in section 2.1.2.

2.1.1 Circular Rydberg States

Circular states have highest electron orbital angular momentum (l) and azimuthal quantum number (m) values within a n manifold i.e. $l = (n - 1)$ and $|m| = (n - 1)$, respectively. Hulet and Kleppner¹², in 1983, were the first to realise circular atoms through a process called "adiabatic microwave transfer method" (AMTM). Later, Delande and Gay¹³, in 1988, proposed a method involving adiabatic switching of crossed fields. This method works on any atomic species. Both methods are capable of producing circular states with values of n up to ~ 60 ¹⁴.

Circular states do not exhibit first-order Stark energy shifts. They have the longest radiative lifetimes ($\tau \propto \mathbf{n}^5$) and are difficult to ionise due to low oscillator strength coupling to the continuum for high l values¹⁵. The selection rules for dipole transitions require that $\Delta l = \pm 1$ and $\Delta m_l = \pm 1$, and therefore only allow spontaneous emission from one circular

state to the next-lower-lying circular state, *i.e.*, by a $\Delta n = -1$ transition. This is useful for many studies as the dynamics of the excitation and decay processes can be written as a two-level system.

Circular atoms are of interest mainly because of their applications in metrology (e.g., a precise determination of the Rydberg constant), QED tests, astrophysics and nowadays in quantum computing research.

2.1.2 Quantum Defect

For completeness, here I discuss the concept of the quantum defect which extends the relations derived to atomic hydrogen (see, e.g., Table 2.1) to the Rydberg states of other atoms.

The quantum defect is a correction factor that was introduced to account for coulomb screening of core electrons while calculating energy eigenvalues of Rydberg states. The table 2.1 shows dependence of several properties on $\mathbf{n}$. This is the effective principal quantum number and is expressed by $\mathbf{n} = n - \delta_{nl}$. The quantum defect decreases with increasing l but n dependence is very small. It is also proportional to the nuclear charge. Note that the quantum defect for hydrogen is zero. Quantum defects are determined experimentally. For large n , δ_{nl} approaches a constant value that depends only on l . The quantum defect is calculated by fitting term energies of different excited states using the **Rydberg-Ritz** formula (Eq 2.2).

$$E_{nl} = E_{\infty} - \frac{E_h}{2(n - \delta_l)^2} \quad (2.1)$$

where E_∞ is the ionisation energy.

$$\delta_{nl} = \delta_l + \frac{B}{(n - \delta_l)^2} + \frac{C}{(n - \delta_l)^4} + \dots \quad (2.2)$$

The table 2.2 summarises quantum defects and ionisation energies for all alkali metals and hydrogen in their ground states¹⁶.

Element	Quantum Defect	Ionisation Energy(eV)
H	0	13.6
Li	0.41	5.4
Na	1.37	5.1
K	2.23	4.3
Rb	3.19	4.2
Cs	4.13	3.9

Table 2.2: Quantum defects, ionisation energies of ground state alkali metals

2.2 Wavefunctions and Eigenvalues of Hydrogen Atom

The spatial wavefunction of the hydrogen atom is given by:

$$\psi_{nlm}(r, \theta, \phi) = \mathcal{R}_{nl}(r)Y_{lm}(\theta, \phi) \quad (2.3)$$

with the radial wavefunction

$$\mathcal{R}_{nl}(r) = \frac{1}{a_0^{3/2}} R_{nl}(r/a_0) \quad (2.4)$$

where $\rho = r/a_0$ and $R_{nl}(\rho)$ is

$$\begin{aligned}
R_{nl}(\rho) &= \frac{2}{n^2} e^{-\frac{\rho}{n}} \sqrt{\frac{(n-l-1)!}{(l+n)!}} \left(\frac{2\rho}{n}\right)^l L_{n-l-1}^{2l+1} \left(\frac{2\rho}{n}\right) \\
&= \frac{2}{n^2(2l+1)!} e^{-\frac{\rho}{n}} \sqrt{\frac{(l+n)!}{(n-l-1)!}} \left(\frac{2\rho}{n}\right)^l \times \\
&\quad {}_1F_1\left(l-n+1; 2l+2; \frac{2\rho}{n}\right). \tag{2.5}
\end{aligned}$$

The angular part is given by the spherical harmonics $Y_m^l(\theta, \phi)$:

$$Y_m^l(\theta, \phi) = (-1)^m \sqrt{\frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!}} P_m^l(\cos \theta) e^{im\phi} \tag{2.6}$$

Using a given Hamiltonian one can evaluate the eigenvalues for different perturbations simply by using the time-independent Schrödinger equation (for static fields) in an appropriate basis.

$$\hat{H}\Psi = E\Psi \tag{2.7}$$

2.3 Effect of Static Electromagnetic Fields on Rydberg Atoms

The splitting of degenerate states in the presence of electric field ($\mathbf{F}$) is known as the Stark effect. The Hamiltonian of Hydrogen in the presence of external electric field can be written as:

$$H_{\text{Total}} = H_{\text{Unperturbed}} + H_{\text{Stark}} \quad (2.8)$$

where the unperturbed Hamiltonian is $H_{\text{Unperturbed}} = \frac{p^2}{2m_e} - \frac{e^2}{4\pi\epsilon_0 r}$ and the Stark Hamiltonian is $H_{\text{Stark}} = e|\mathbf{F}|z$. The Schrödinger equation for the hydrogen atom, $H_{\text{Stark}}\psi(\mathbf{r}) = E\psi(\mathbf{r})$ (in atomic units $\hbar = m = e = 1$), can be separated and solved in parabolic coordinates (ξ, η, ϕ) as well as in the more conventional spherical polar coordinates (r, θ, ϕ) . This indicates existence of a hidden symmetry, the $\text{SO}(4)$ associated with the Coulomb problem. Parabolic coordinates can be defined by $\xi = r(1 + \cos \theta) = r + z$, $\eta = r(1 - \cos \theta) = r - z$, with the same $\phi = \arctan(y/x)$ as in spherical coordinates. The wavefunction is separable in the form $\psi_{n_1 n_2 m}(\xi, \eta, \phi) = N f_1(\xi) f_2(\eta) e^{im\phi}$ with $f_{1,2}(\zeta) = \zeta^{-1/2} M_{n_{1,2} + \frac{|m|+1}{2}, \frac{|m|}{2}}(\zeta/n)$. Here M is a Whittaker function and $n = n_1 + n_2 + |m| + 1$. The hydrogen atom in a constant electric field is also separable in parabolic coordinates. The functions $f_1(\xi)$ and $f_2(\eta)$ are more complicated but can be obtained by perturbation expansions. Probability density of some Hydrogen wavefunctions in parabolic co-ordinates have been presented in fig.2.2. To first order, the Stark effect energies are given by¹⁷:

$$E_{n n_1 n_2} = -\frac{1}{2n^2} + \frac{3}{2} F n (n_1 - n_2) \quad (2.9)$$

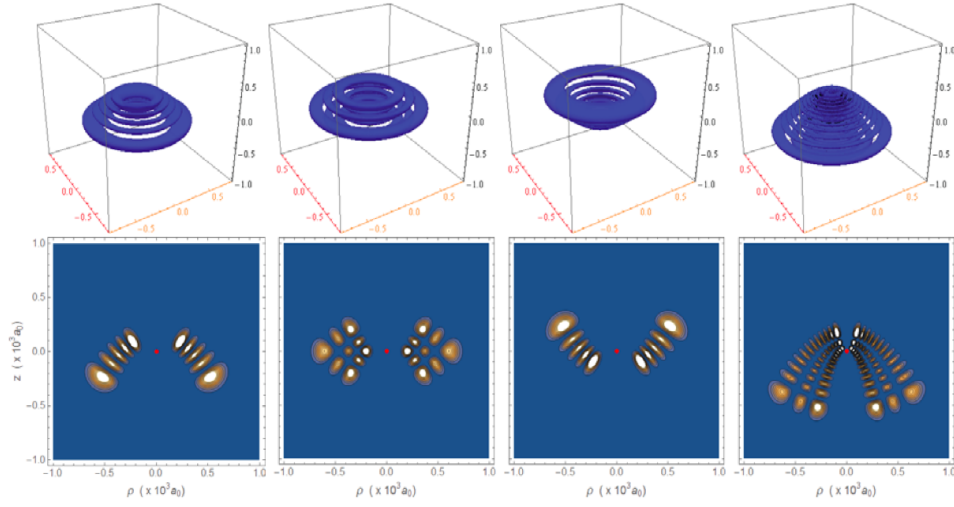


Figure 2.2: Probability density of some Hydrogen wavefunctions in parabolic co-ordinates for $n=21$. From the left: $|n_1, n_2, m\rangle = |0, 4, 16\rangle, |2, 2, 16\rangle, |4, 0, 16\rangle, |2, 10, 7\rangle$. In the first three plots n_1, n_2 is changed keeping m constant. Plots adapted from ref. ¹⁸

It can be observed from Eq. 2.9 that the energy of eigenstates is dependent on the combination of the two parabolic numbers n_1, n_2 . ($k = n_1 - n_2$) is called as the electric quantum number. Notice that m does not appear in the first order Stark effect, it only appears in higher orders. One atomic unit of electric field $\mathbf{F}$ is equivalent to 5.142×10^{11} V/m. The two outermost components are the: $(n_1, n_2) \rightarrow (n-1, 0)$ and $(0, n-1)$, the first one being blue shifted and the second one being red shifted (see $n=8, k = +7, -7$ states in ref. fig. 2.3). Their energy difference is

$$\Delta E = 3Fn(n-1) \quad (2.10)$$

It can be observed that the energy difference between the outermost states grows proportional to n^2 . In a Plasma, there exist fluctuating fields generated by the surrounding charged particles. This leads to distortion of the energy of states. At a certain intensity of electric field, the stark states in n manifolds start "mixing" and cross with the neighbouring n manifolds (crossings can be seen in fig.2.3 (a)). In non-Hydrogenic atoms avoided crossing occur due to the presence of quantum defects. The intensity of the electric field where the

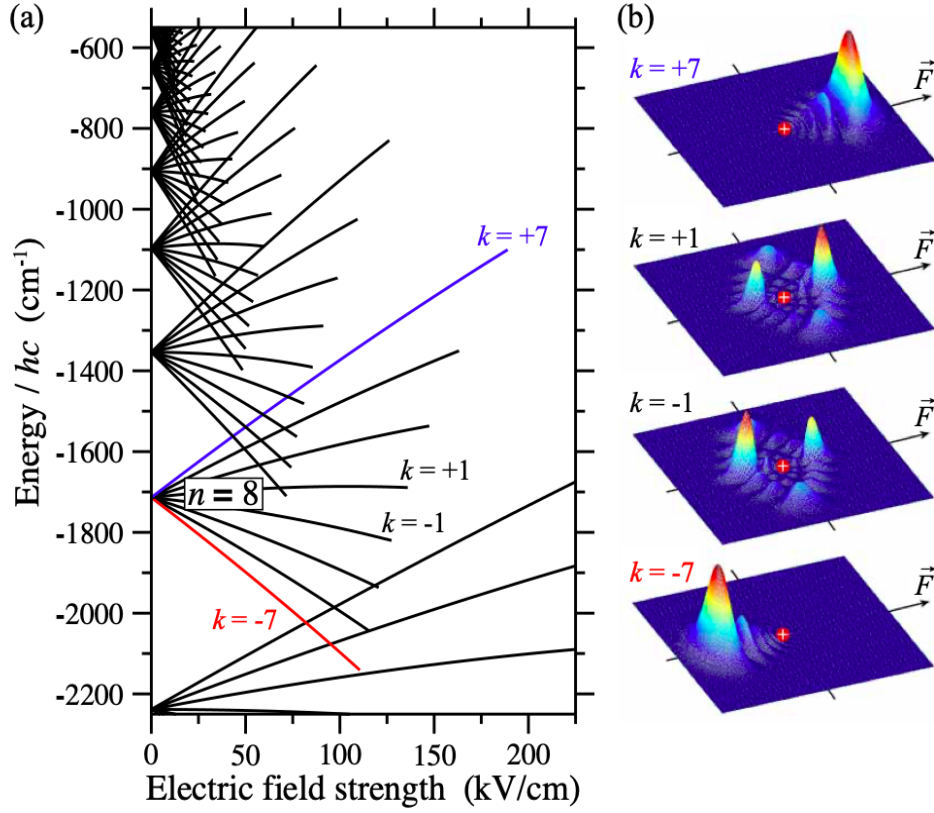


Figure 2.3: The Stark effect in Rydberg states of H. a) Stark energy splittings in $n = 7 - 14$ manifolds at $m=0$. b) The probability density of electron of $n=8$ for values of $k = n_1 - n_2 = +7, +1, -1, -7$ along with the orientation of $\vec{F}$ field axis. Adapted from ref.¹⁹

intra- n manifold mixing begins is known as the Inglis-Teller limit, given by Eq. 2.11 (in atomic units).

$$F_{\text{Ing.Tel.}} = \frac{1}{3n^5} \quad (2.11)$$

An applied magnetic field also produces perturbations in eigenenergies due to its interaction with the magnetic moment of the atom.

$$H_{\text{Total}} = H_{\text{Unperturbed}} + H_{\text{Zeeman}} \quad (2.12)$$

$H_{\text{Zeeman}} = -\boldsymbol{\mu} \cdot \mathbf{B}$ where $\boldsymbol{\mu} = -\frac{\mu_B}{\hbar}(g_L \mathbf{L} + g_S \mathbf{S})$. $\mu_B = \frac{e\hbar}{2m_e}$ is the Bohr magneton and $\mathbf{L}, \mathbf{S}$ are the orbital angular momentum and the spin angular momentum respectively. g_L and g_S are the gyromagnetic ratios. Then, the Zeeman perturbation can be written as $H_{\text{Zeeman}} = \frac{\mu_B B}{\hbar}(g_L \mathbf{L} + g_S \mathbf{S})$. The eigenvalues of H in presence of magnetic field are shifted in the parabolic coordinates by:

$$\Delta E_{\text{Zeeman}} = \langle n_1 n_2 m m_s | H_{\text{Zeeman}} | n_1 n_2 m m_s \rangle = \mu_B B (g_L m + g_S m_s) \quad (2.13)$$

where m_s denotes the spin quantum number.

Now that we have a foundation on the effects of electric and magnetic field on H in parabolic coordinates, the technique to produce circular state atoms can be discussed.

The idea is to excite electron to a low m value like $m=0$ using laser in the Stark limit (when $\mathbf{F} \gg \mathbf{B}$) and adiabatically switch off the $\mathbf{F}$ field while holding the small $\mathbf{B}$ field constant. The adiabaticity condition is given in Eq. 2.14. The outermost stark states within a n manifold are those of $m = 0$ i.e., $(n_1, n_2) \rightarrow (n-1, 0)$ and $(0, n-1)$. These states transform into the $m = n-1$ circular state (in spherical basis) after the $\mathbf{F}$ field is adiabatically turned off.

$$\dot{F} \ll \frac{18B^2}{n} \quad (2.14)$$

In this equation, $\dot{F}$ is given in V/m/ μs and B is given in G. With this condition, for a $\mathbf{B}$ field of tens of Gauss, the $\mathbf{F}$ field has to be switched off within tens of μs . If this condition is satisfied then it results in the transformation of the $m=0$ state into the $|m| = n-1$ circular state. An excitation scheme can be seen illustrated in terms of an energy diagram in fig. 2.5 to produce $n=29$ circular state in Hydrogen. A similar example has been shown in fig. 2.4

for $n=5$ manifold of Hydrogen. This method is known as the crossed fields method (CFM). It has been consistently proven to be $\sim 100\%$ efficient. For more details on the CFM, see ref.²⁰ by Lutwak et al.

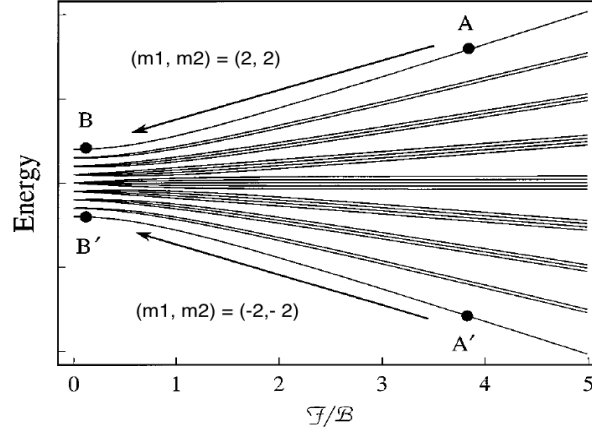


Figure 2.4: Energy levels of the hydrogen $n=5$ manifold in nearly perpendicular electric and magnetic fields. The $\mathbf{F}$ and $\mathbf{B}$ fields are oriented at $\sim 100^\circ$. A, A': initial states. B, B': final, circular states. Plot reproduced from ref.²⁰, fig. 1.

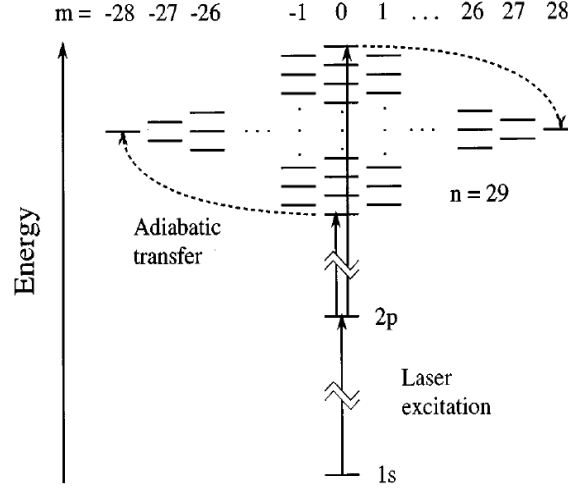


Figure 2.5: Circular state excitation scheme for $n=29$ in atomic Hydrogen. Plot adapted from ref.²⁰, fig. 5.

Chapter Three

Simulation: Excitation of Hydrogen to Rydberg Circular State

The presented numerical simulation aims to find optimum values for the strength of electric and magnetic fields and their mutual orientation that optimally mix states and produce maximum transition rates from 2s to the $n=30$ circular state. Einstein, in 1923, was the first physicist to consider first-order perturbation theory for the hydrogen atom in crossed electric and magnetic fields with a classical approach. Later, Demkov et. al.^{[21](#)}, in 1969, developed a quantum mechanical approach to find energy splittings using first and second-order perturbation theory.

In this simulation, a simplified model has been used that does not calculate the energy splittings by diagonalising the Stark and the Zeeman perturbation Hamiltonians in the spherical basis introduced in Sec. [2.2](#) but by considering a specific basis which already diagonalises the perturbing operators. Calculating transition rates to the circular state then reduces to evaluating the overlap between basis vectors. This takes only the first-order energy perturbations into account to estimate some quantities like the amount of Stark and Zeeman splitting and to calculate values of quantum numbers m_1, m_2 in certain regimes. The

theoretical background can be found in section 3.1. The simulation is discussed in section 3.2. Briefly, the simulation methodology is as follows: At first, the radial wave functions of the hydrogen atom in the spherical basis are defined. Then, the states in spherical basis are transformed into the so-called "Lenz-Runge basis" basis using Clebsch-Gordan coefficients and real Wigner-D matrices. By applying the Wigner-Eckart theorem, it is possible to calculate the dipole matrix elements between the 2s and the $n=30$ circular state. Finally, the transition rate, being directly proportional to the square of the reduced dipole matrix element, is plotted for different values of the electric field $\mathbf{F}$ and the angle between electric and magnetic fields. The value of the magnetic field strength $\mathbf{B}$ is always kept constant. Transition rates for different polarisations of the laser light are considered.

The equations used in this chapter have been adapted from Ref.⁸ unless stated otherwise.

3.1 Theoretical Background for the Simulation

The Hamiltonian of the hydrogen atom in crossed external fields can be written as

$$H = -\frac{1}{2n^2} + V_1 + V_2 \quad (3.1)$$

where the perturbation V_1 is given by $V_1 = \mathbf{r} \cdot \mathbf{F} + \frac{1}{2}\mathbf{B} \cdot \mathbf{L}$. Only the first order perturbation is considered, as the contribution from the second order term $V_2 = -\frac{1}{8}(\mathbf{r} \times \mathbf{L})^2$ is on the order of $n^6 \mathbf{F}^2 + n^4 \mathbf{B}^2/4$ ²² and is negligible compared to the first order for our purposes. Throughout this chapter, electric fields $\mathbf{F}$ and magnetic fields $\mathbf{B}$ have been denoted in atomic units, the conversion factors being $\mathbf{F}_0 = 5.14 \times 10^{11}$ V/m and $\mathbf{B}_0 = 2.35 \times 10^5$ T. In the presence of small enough $\mathbf{F}$ and $\mathbf{B}$ fields that do not produce second-order effects, only the z components of Lenz-Runge vector $\mathbf{A}_z$ and the classical angular momentum vector $\mathbf{L}_z$

are conserved. That was shown by Pauli²³. As Pauli has shown, the vector $\mathbf{r}$ is proportional to the Lenz-Runge vector $\mathbf{A}$ as $\mathbf{r} = -\frac{3}{2}n\mathbf{A}$ for a given n manifold. We can define two commuting angular momenta $\mathbf{I}_1$ and $\mathbf{I}_2$:

$$\mathbf{I}_1 = \frac{\mathbf{L} + \mathbf{A}}{2}; \mathbf{I}_2 = \frac{\mathbf{L} - \mathbf{A}}{2}; I_1 = I_2 = \frac{n-1}{2}. \quad (3.2)$$

With the two quantization axes

$$\boldsymbol{\omega}_1 = \frac{\mathbf{B} - 3n\mathbf{F}}{2}; \boldsymbol{\omega}_2 = \frac{\mathbf{B} + 3n\mathbf{F}}{2} \quad (3.3)$$

one can define a new basis $|I_1 m_1\rangle \otimes |I_2 m_2\rangle$, where $m_1, m_2 = -(n-1)/2, -(n-3)/2, \dots, (n-1)/2$. This basis is called the Lenz-Runge basis. The two new quantum numbers m_1, m_2 are the projections of $\mathbf{I}_1$ and $\mathbf{I}_2$ on $\boldsymbol{\omega}_1$ and $\boldsymbol{\omega}_2$. With the help of Eq. 3.2 we write the first order perturbation as $V_1 = \boldsymbol{\omega}_1 \cdot \mathbf{I}_1 + \boldsymbol{\omega}_2 \cdot \mathbf{I}_2$. The basis $|I_1 m_1\rangle_{\boldsymbol{\omega}_1} |I_2 m_2\rangle_{\boldsymbol{\omega}_2} = |nm_1 m_2\rangle$ diagonalises the perturbing Hamiltonian. The first order perturbation energy is given by

$$\Delta E_{m_1 m_2}^{(1)} = \omega_1 m_1 + \omega_2 m_2. \quad (3.4)$$

For a pure electric or magnetic field we restore the pure Stark effect

$$\Delta E_{m_1 m_2}^{(1)} = \frac{3}{2}nF(m_1 + m_2) \quad (3.5)$$

or Zeeman effect

$$\Delta E_{m_1 m_2}^{(1)} = \frac{1}{2}B(m_1 + m_2), \quad (3.6)$$

respectively.

3.2 Numerical Simulation

The relation among Clebsch-Gordan coefficients, the Runge-Lenz basis and the spherical basis is given by

$$|nlm\rangle_B = \sum_{m_1 m_2} C_{I_1 m_1, I_2 m_2}^{lm} |I_1 m_1\rangle_B |I_2 m_2\rangle_B \quad (3.7)$$

and

$$|I_1 m_1\rangle_B |I_2 m_2\rangle_B = \sum_{ml} C_{I_1 m_1, I_2 m_2}^{lm} |nlm\rangle_B = \sum_l C_{I_1 m_1, I_2 m_2}^{l, m_1+m_2} |nl, m = m_1 + m_2\rangle_B. \quad (3.8)$$

The subscript B indicates that the quantization axis is along B .

Now, we define two angles, α_1 and α_2 , between the magnetic field B axis and the vectors ω_1 and ω_2 respectively. With the help of the Wigner D rotation matrix, we can derive the equation²¹

$$|I_1 m_1\rangle_{\omega_1} = \sum_{m'_1=-I_1}^{m'_1=I_1} D_{m_1, m'_1}^{I_1}(0, \alpha_1, 0) |I_1 m'_1\rangle_B. \quad (3.9)$$

The real Wigner (small) d -matrix is given by

$$d_{m_2, m'_2}^{I_2}(\alpha_2) = D_{m_2, m'_2}^{I_2}(0, \alpha_2, 0). \quad (3.10)$$

By combining Eqs. 3.7, 3.8 (using only real Clesch-Gordan coefficients), 3.9, and 3.10 we get:

$$|nm_1 m_2\rangle_{\omega_1, \omega_2} \equiv |I_1 m_1\rangle_{\omega_1} |I_2 m_2\rangle_{\omega_2} = \sum_{l=0}^{n-1} \sum_{m=-l}^l \langle nlm | nm_1 m_2 \rangle |nlm\rangle_B, \quad (3.11)$$

$$\langle nlm | nm_1 m_2 \rangle \equiv \sum_{m'_1=-I_1}^{m'_1=I_1} \sum_{m'_2=-I_2}^{m'_2=I_2} d_{m_1, m'_1}^{I_1}(\alpha_1^{(n)}) d_{m_2, m'_2}^{I_2}(\alpha_2^{(n)}) C_{I_1 m'_1, I_2 m'_2}^{lm}, \quad (3.12)$$

Equation 3.11 shows that the coupled states can be expanded via the completeness relation in the uncoupled basis. Here, the expansion coefficient $\langle nlm|nm_1m_2\rangle$ contains the Clebsch-Gordan coefficient. Equation 3.12 was derived by Park²⁴ exploiting the fact that the Runge-Lenz vector and angular momentum are conserved in hydrogen in zero field and he showed that the transformation coefficient between the parabolic and spherical basis is just a Clebsch-Gordan coefficient. Here, $\alpha_1^{(n)}$ indicates that the angles depend not only on the fields' values but also on n . We can simplify Eq. 3.11 by writing it as $|nlm\rangle_{\mathbf{B}} = \sum_{m_1,m_2} \langle nm_1m_2|nlm\rangle |nm_1m_2\rangle_{\omega_1,\omega_2}$ and then use the closure expression, *i.e.* $\mathbf{1}_n = \sum_{m_1,m_2} |nm_1m_2\rangle \langle nm_1m_2| = \sum_{lm} |nlm\rangle_{\mathbf{B}\mathbf{B}} \langle nlm|$, that can be used in both the spherical and parabolic bases.

The cross-fields method to prepare atoms in circular states (ref.²⁰) involves excitation of $m = 0$ Rydberg state along a quantization axis and then adiabatically redefining it perpendicularly into $|m| = n - 1$. This state is always the outermost state of the split manifold. In a pure electric field, $k = -(m_1 + m_2)$, $m = m_2 - m_1$ (ref. Eqs. 3.4, 3.5) and for the circular state $k = 0$, $|m| = n - 1$. So we get $m_1 = -m_2 = \frac{n-1}{2}$ that leads, for $n = 30$, to two solutions for the most strongly shifted state, *i.e.* $m_1, m_2 = [(\frac{29}{2}, -\frac{29}{2}), (-\frac{29}{2}, \frac{29}{2})]$. In a pure magnetic field, $m = m_2 + m_1$ (ref. Eqs. 3.4, 3.6) and for circular state in crossed fields $|m| = n - 1$. So we get $m_1 = m_2 = \frac{n-1}{2}$ that leads to two solutions, *i.e.* $m_1, m_2 = [(\frac{29}{2}, \frac{29}{2}), (-\frac{29}{2}, -\frac{29}{2})]$. By equating Eqs. 3.5 and 3.6 we find the condition for equal Stark and Zeeman splitting, which is $\frac{B}{F} = 3n$. When F exceeds this ratio then the Stark splitting dominates over the Zeeman splitting so the m_1, m_2 values change for the outermost state. When Eq. 3.4 is plotted against F , it is observed that the linear energy splitting starts at the critical value of $F = \frac{B}{3n}$. So the state that becomes the circular state at zero electric field has the quantum numbers $m_1 = -m_2 = \frac{n-1}{2}$ above this critical field value while below this field, it corresponds to the state with the quantum numbers $m_1 = m_2 = \frac{n-1}{2}$.

The dipole matrix elements can be easily evaluated with the obtained m_1m_2 values.

Thus the transition rate between $|30m_1m_2\rangle$ and the 2s state can be plotted using

$$\begin{aligned}\Gamma_{\text{Absorp}}^{nm_1m_2;n';q} &= \frac{2Ie^2a_0^2}{\hbar^2\epsilon_0c\Gamma_L} \sum_{l',m'} \left| \langle n'l'm' | \frac{r^{(q)}}{a_0} | nm_1m_2 \rangle \right|^2 \\ &= \frac{2Ie^2a_0^2}{\hbar^2\epsilon_0c\Gamma_L} \sum_{m,l'} \left| \sum_{l=l'\pm 1} \langle n'l'm+q | \frac{r^{(q)}}{a_0} | nlm \rangle \langle nlm | nm_1m_2 \rangle \right|^2\end{aligned}\quad (3.13)$$

where $q = \Delta m = \pm 1, 0$ indicates the polarisation of the driving laser light. This equation can be evaluated using the dipole matrix elements between different states, given by the Wigner-Eckart theorem

$$\begin{aligned}\langle n'l'm' | r^{(q)} / a_0 | nlm \rangle &= C_{lm,1q}^{l'm'} \frac{\langle n'l' || r / a_0 || nl \rangle}{\sqrt{2l'+1}} \\ &= C_{lm,1q}^{l'm'} C_{l0,10}^{l'0} \frac{\sqrt{2l+1}}{\sqrt{2l'+1}} R_{nl}^{n'l'}.\end{aligned}\quad (3.14)$$

This expression contains only Clebsch-Gordan coefficients and the radial overlap term $R_{nl}^{n'l'} = \int_0^\infty R_{nl}(\rho) \rho R_{n'l'}(\rho) \rho^2 d\rho = \mathcal{R}_{nl}^{n'l'} / a_0$, which is given in the atomic units by

$$\begin{aligned}R_{nl}^{n'l-1} &= \frac{(-1)^{n'-l}}{4(2l-1)!} \sqrt{\frac{(l+n)!(l+n'-1)!}{(-l+n-1)!(n'-l)!}} \frac{(4nn')^{l+1}(n-n')^{n+n'-2l-2}}{(n+n')^{n+n'}} \\ &\quad \left({}_2F_1 \left(l-n+1, l-n', 2l, -\frac{4nn'}{(n-n')^2} \right) \right. \\ &\quad \left. - \frac{(n-n')^2}{(n'+n)^2} {}_2F_1 \left(l-n-1, l-n', 2l, -\frac{4nn'}{(n-n')^2} \right) \right).\end{aligned}\quad (3.15)$$

The experimental parameters and values of physical constants used in the calculation of transition rates are given in Tab. 3.1.

Constant	Description	Value
I^1	Max power density of the seeded beam	3.18 W/cm ²
Γ_L	Linewidth of laser	~ 100 MHz
a_0	Bohr radius	0.529×10^{-10} m
e	Charge of electron	1.602×10^{-19} C
ϵ_0	Bohr radius	8.854×10^{-12} F/m
c	Speed of light	2.996×10^8 m/s
$\hbar$	Planck's constant	6.626×10^{-34} Js

Table 3.1: Table of laser parameters and constants. The seeded beam (366 nm) has a temporal and spatial Gaussian beam profile with pulse duration (6 ns), avg. pulse energy (20 mJ), and beam radius (2 mm). The repetition rate is 10 Hz. The peak power density was calculated using [Ophir's calculator](#).

3.3 Results of the Simulation

The simulation was performed using **Wolfram Mathematica 12.3.1**²⁵. The calculations have been done within the first-order approximation, so the lower limit of the transition rates is at the point where the Stark shift just exceeds the Zeeman shift (at $F = \frac{B}{3n}$ and above). The Inglis-Teller limit for $n=30$ manifold was also considered to limit the calculations. It turns out to be $F_{\text{Ing.Teller}} \approx 7120$ V/m (ref. Eq. 2.11).

Here, I choose two values of the magnetic flux density (20 G, 100 G). These values have been chosen by referring to previous studies done on the crossed fields method^{13,20,26}. The simulation results can be summarised as follows:

1. The highest transition rates occur near perpendicular field orientations. But, it should be noted that within the first order perturbation theory, if $\mathbf{F} \cdot \mathbf{B} = 0$, then the quantization axes $\omega_1 = \omega_2$ coincide. It means that the degeneracy is not completely removed. So for maximum mixing, the fields should have a slight angular deviation of $\sim 10^\circ$ ²⁰ from perpendicular orientations.
2. The transition rates depend on the ratio of the Stark/Zeeeman splitting rather than

pure field strengths. The optimal fields are related through the relation $3nF \sim B$ in atomic units ($B/(2.35 \times 10^5 \text{ T})$ and $F/(5.14 \times 10^9 \text{ V/cm})$). The optimal F field values for chosen B values are ($\sim 140 \text{ V/m}$, 20 G) or ($\sim 700 \text{ V/m}$, 100 G).

3. The maximum transition rates for both values of the B field are of the order of 10^{13} s^{-1} for the chosen parameters of the $\sim 366 \text{ nm}$ beam (see table 3.1). The rates are practically independent of the beam polarisation.
4. The maximum transition rates barely change if plotted upto the Inglis-Teller limit 7120 V/m (here, I do not plot upto the Inglis-Teller limit). So, electric fields as low as $\sim 1.5 \text{ V/cm}$ and $B = 20 \text{ G}$ should suffice to mix states and excite electrons to the outer states of the $n=30$ manifold. Higher F fields can also be used.
5. An important thing to consider is the bandwidth of the laser. Transfer of the electron to the outermost Stark state (in the Stark limit) depends on the amount of splitting produced by the $\mathbf{F}$ field between neighbouring Stark states and the bandwidth of the laser. Our laser has a bandwidth of $\sim 100 \text{ MHz}$. The splitting between neighbouring stark levels is calculated by $\Delta E_{\text{neigh.lev.}} = \sqrt{\omega_{\text{Stark}}^2 + \omega_{\text{Zeeman}}^2}$ where $\omega_{\text{Stark}} = 3nF/2$ and $\omega_{\text{Zeeman}} = B/2$ are the Stark and Larmor frequencies. For $\sim 170 \text{ V/m}$ and 20 G , $\Delta E_{\text{neigh.lev.}} \approx 100 \text{ MHz}$. So, an electric field above $\sim 170 \text{ V/m}$ would ensure selective excitation to the outermost Stark states.

The optimal fields found are feasible to implement in the experiment. A pair of Helmholtz coils can easily produce a 20 G magnetic field, a pair of electrodes to which modest potentials are applied can produce the electric field. Practically, implementing fields as low as $F < 200 \text{ V/m}$ is a challenge since a stray field of the order of 100 V/m is present in the excitation region. By applying high fields of about $3000\text{-}5000 \text{ V/m}$, the stray field is significantly exceeded, and therefore the angle between the magnetic and the electric field can be defined more precisely and thus, it is easier to implement. This magnitude of F field

will be ideal for selective excitation of states as $\Delta E_{neigh.lev.}$ will be about 1700-2800 MHz in this range.

Apart from applying fields and exciting the atoms with a laser, another step is required to successfully produce circular state atoms. The criteria is that either of the fields must be adiabatically switched off while holding the other field constant (ref.²⁰). The adiabaticity condition is given by Eq. 3.16.²⁰ For $F = 140$ V/m, $B = 20$ G, the time in which the F field should be turned off turns out to be in a few or tens of μ s. If followed, it would lead to 100% efficient circular state production.

$$\dot{F} \ll \frac{18B^2}{n} \quad (3.16)$$

In this equation, $\dot{F}$ is given in V/cm/ μ s and B is given in G.

Angular momentum l	Spontaneous lifetime τ (ms)	Velocity of H (m/s)
29	2.32	216
20	1.12	446
13	0.49	1030
6	0.113	4440
4	0.054	9300
1	0.006	83000

Table 3.2: Lifetimes of various l states of H $n = 30$ manifold and the minimum velocity of H required for the respective states to be detectable before they decay to GS. The velocities were calculated considering an approximate distance of 0.5 m between the detection region and the H source.

With the given adiabaticity condition in Eq. 3.2, it can be evaluated that if $B = 20$ G, $F = 3000 - 5000$ V/m then an F field has to be adiabatically turned off in 10μ s (or slower). The electric field has to be implemented starting at the point of excitation and adiabatically ramped down in 5 cm to produce and detect circular states. We expect the Hydrogen atoms from the plasma source to have velocities of a few 1000 m/s. With the expected velocity and spontaneous lifetimes as given in table 3.2, we can expect to detect states having $l = 6$ to

$l = 29$. The B field has to be held approximately constant throughout the ~ 0.5 m region.

Simulation: Excitation of Hydrogen to Rydberg Circular State

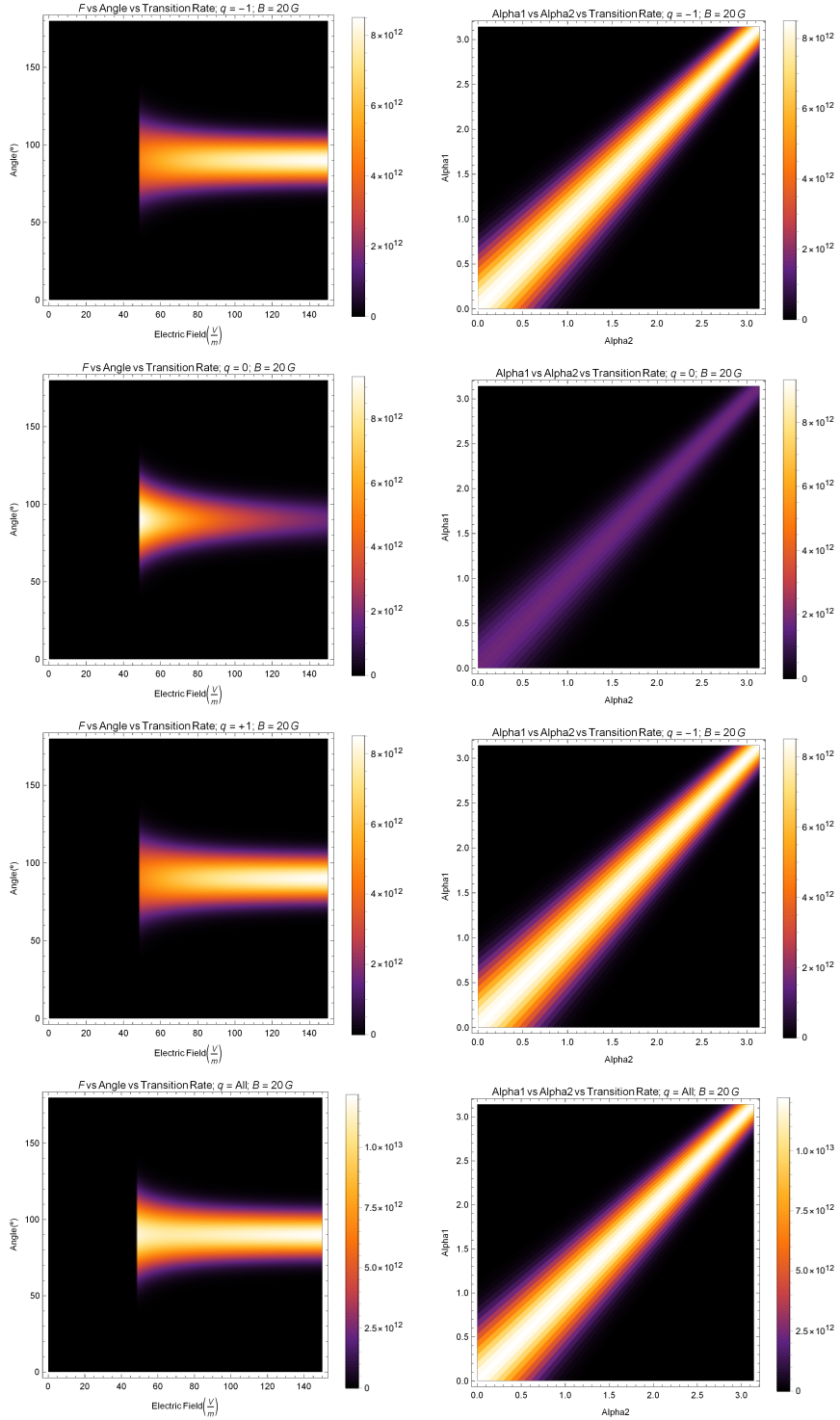


Figure 3.1: Transition Rate Plots for $B=20$ G. First column: transition rates vs. electric field and angle. Second column: transition rates vs. α_1 and α_2 . From top to bottom $q = (-1, 0, +1, \text{All})$ respectively. Maximum rates are observed near 90° orientation of fields and at higher electric fields (140 – 150 V/m).

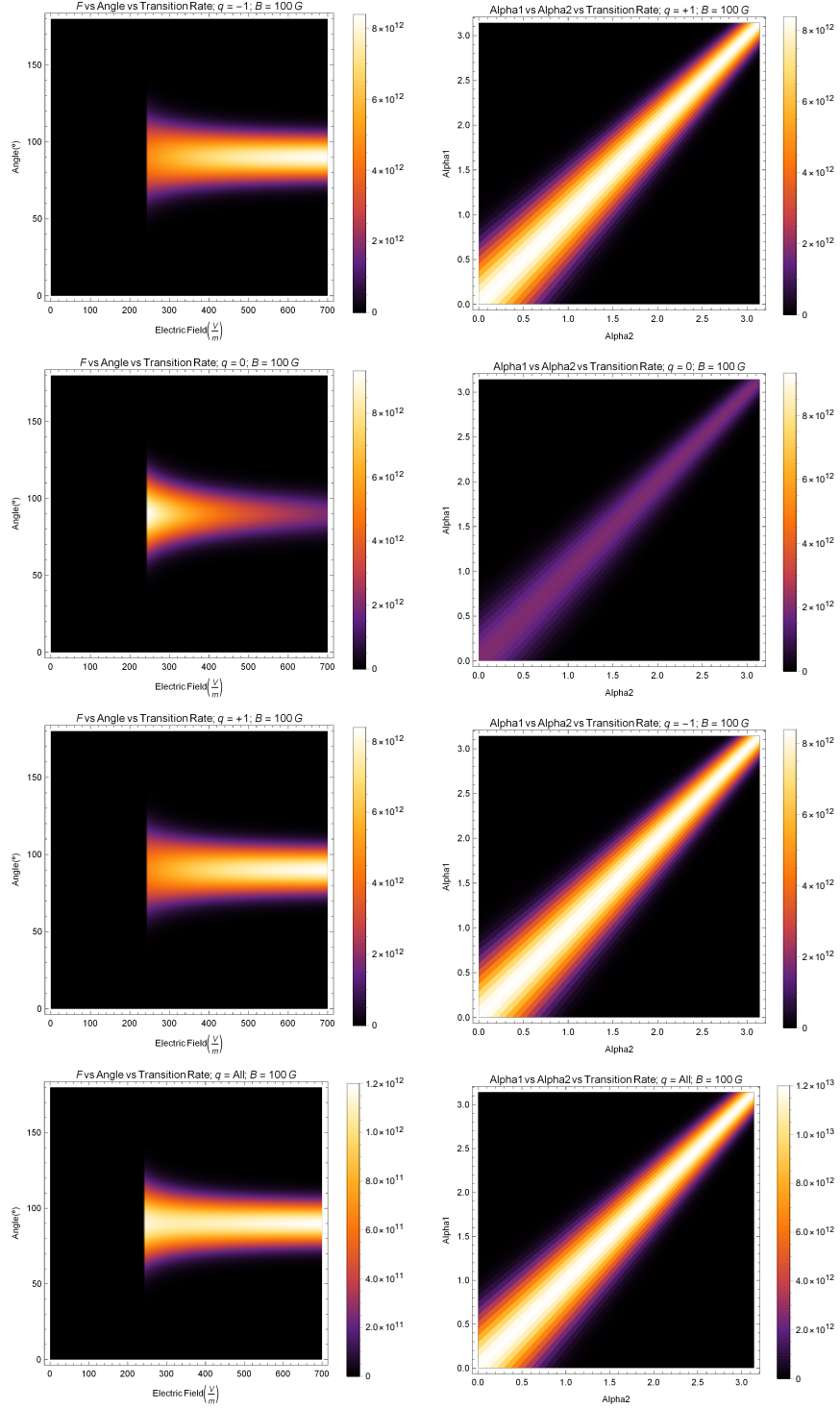


Figure 3.2: Transition rate plots for $B=100$ G. First column: transition rate vs. electric field and angle. Second column: transition rates vs. α_1 and α_2 . From top to bottom $q=(-1, 0, +1, \text{All})$ respectively. Maximum rates are observed near 90° orientation of fields and at higher electric fields (600 – 700 V/m).

Chapter Four

The Hydrogen Excitation Experiment

This chapter presents the construction and characterisation of the experimental setup to excite H from 2s to 30p. First, in section 4.1, an overview of the setup is given. Then, in section 4.2, the developments in the apparatus are discussed in four subsections. First, in subsection 4.2.1, the results of the spectroscopic measurements of the plasma are analysed. Next, in subsection 4.2.2 and 4.2.3, the details of beam production, excitation of H and detection of Rydberg states are discussed. Then, in subsection 4.2.4, the visualised doppler measurements to estimate the velocity of the outcoming H atoms and the details of the diode laser used is discussed. Lastly, in subsection 4.2.5, the details of a custom LabView VI programmed to acquire experimental data are explained. In this experiment, my contributions were the following:

1. **Installation of the beamline:** I helped install the beamline, then integrated different instruments like the turbopump, cold head, plasma cavity, flow meter, microwave source, etc. I helped build and fit a laser transfer bridge from the laser table to the beamline.
2. **Programming LabVIEW VI:** I programmed a LabVIEW VI to integrate sensors with the PC to control them individually and acquire data.

3. **Spectroscopic measurements:** I took spectroscopic measurements to characterise the plasma source and to find the intensities of the H_α, H_β transitions. Before taking measurements, I calibrated the spectrometer.
4. **Setting up and calibration of the lasers:** I contributed to setting up the Nd:YAG pump laser, optimising its output power, tuning the wavelength of the diode laser, tuning the Ti:Sa cavity.
5. **Troubleshooting devices:** I worked on troubleshooting wavelength stability of a diode laser by inspecting a laser diode controller, temperature controller, and issues with the powermeter.
6. **Miscellaneous:** I soldered electronic circuits, periodically documented experimental details, tracked stability of pressure and temperature in the beamline.

4.1 Overview of the Setup

Figure 4.1 shows a labelled photograph of the experiment. The setup consists of a laser setup and a beamline. For better understanding, schematic diagrams of the laser setup (ref. fig.4.9) and the beamline (ref. fig.4.11) are also included.

On the laser table the light needed to produce the $2s$ to $n=30$ excitation is generated. It starts from a pulsed commercial Nd:YAG (T1) which is frequency doubled to ~ 532 nm and injected into a gain switched Ti:Sa (T3) cavity seeded by a diode laser (T2) at ~ 732 nm. This beam is frequency doubled to ~ 366 nm using a BBO crystal (T5). Then the beam is passed through a transfer stage to the excitation region in the beamline (C2).

In the beamline, starting from the left, is an Evenson cavity (C1) where the plasma

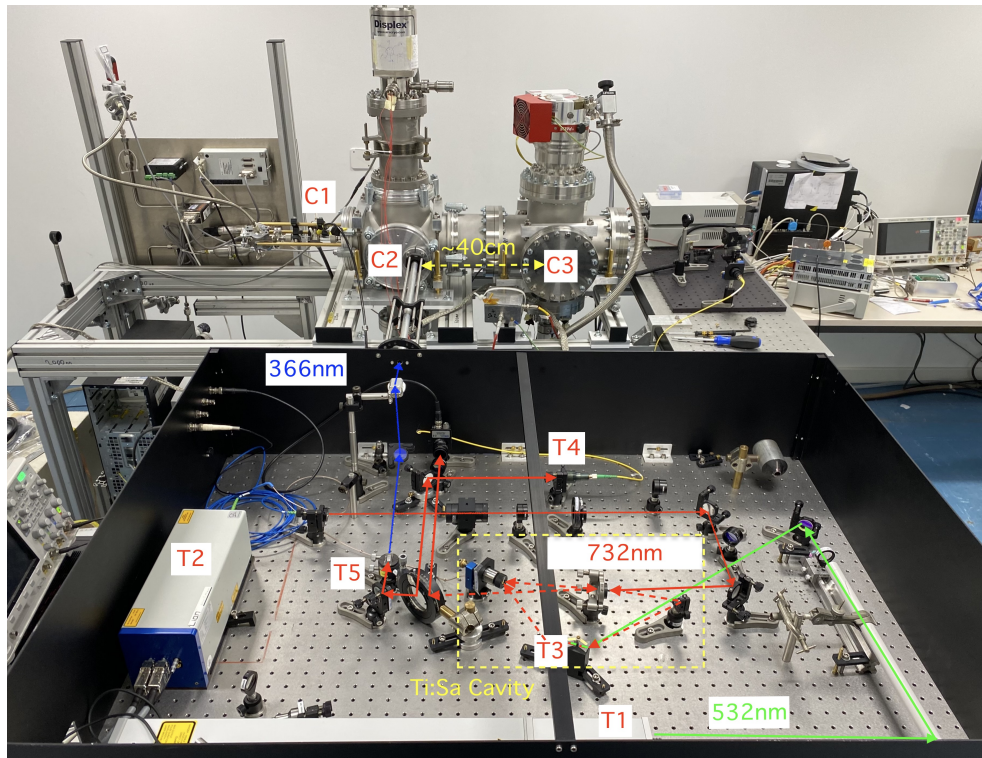


Figure 4.1: Photograph of the Experimental Setup: T1-Pulsed Nd:YAG laser, T2-Diode laser for seeding, T3-Ti:Sa crystal (behind the bar), T4-Fiber coupler (to the wavemeter), T5-BBO Doubling crystal, C1-Evenson cavity, C2-Excitation region C3-Detection region. Arrows indicate beam paths and the coloured lines have respective wavelengths indicated in the same colour.

is produced. This source is capable of producing excited states of atomic Hydrogen (yet to be quantified). Among the excited states is the 2s state which we envision to excite to $n=30$ circular state. After the atoms arrive in the excitation region (C2) they will be excited by the ~ 366 nm laser beam. Then they will fly through a thermally shielded passage (ref. fig. 4.11). The thermal shielding is a cooled copper structure to shield the Rydberg atoms from blackbody photons at 300K. It is a two stage shield cooled to ~ 40 K with a cryocooler. Finally, the excited atoms fly to the detection region (to the chamber at the top in fig. 4.11) where the field ionisers would ionise the Rydberg H atoms. Field ionisers (FI) consist of two metallic grids connected to high voltage supplies that generate an electric field between the two plates. By varying the voltage on the FI we can selectively ionise different states. Microchannel plates (MCPs) situated on the either sides of the beamline are used to detect

the particle count.

It should be noted that the experiment is still under development. The next section informs about the current developments in the apparatus and the details associated.

4.2 Developments in the Apparatus

4.2.1 Hydrogen Production and Characterisation of the Source

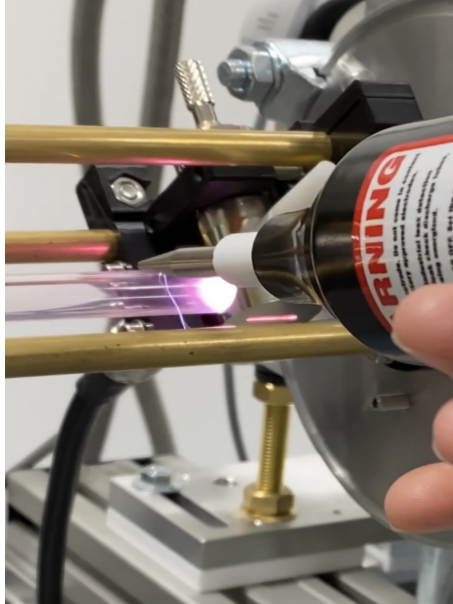


Figure 4.2: The picture shows a Hydrogen plasma being ignited with a spark gun in an Evenson microwave cavity that operates at 2450MHz. The cavity can be tuned using two knobs to optimise the reflected power. Balmer- H_α , H_β are the dominant transition lines in the plasma.

We require H atoms in 2s state for our experiment to produce $n=30$ circular state atoms. Many 2s states are produced within the plasma discharge source. We aimed to measured the amount of 2s released in the beam to evaluate if it is sufficient for the needs of the experiment. The reason to excite from the 2s H is that the energy difference between 1s and 2s state is large (corresponding to $\lambda = 121.5$ nm). In case the amount of 2s in the

beam emerging from the source turns out to be too small, several alternatives have already been investigated by the group (e.g. electron impact excitation). Laser excitation from ground-state to 2p is also possible but requires a Lyman-alpha laser.

Hydrogen gas (H_2) from a Hydrogen generator enters an Evenson microwave cavity in which a microwave generator produces radiation of 2450 MHz. A MW source is not enough to ignite the plasma. An external power source has to be coupled to the plasma. A spark from an electric gun induces electric discharge in the presence of microwaves which leads to ignition of the plasma (ref. fig. 4.2). The MW frequency is resonant in the Evenson cavity. It means that at 2450 MHz, standing EM waves are formed within the cavity which results in very low losses and leads to a high Q-factor. Thus more microwave photons are confined within the cavity walls, and the efficiency of plasma ignition is higher. The type of plasma we produce is known as a recombining plasma (that has electron temperature $T_e < 3\text{ eV}$) as recombination processes are dominant in them.

With field ionisation techniques we have found traces of a variety of Rydberg states coming out of the plasma indicating possible existence of 2s population. As of now, the population densities of states are not known.

To quantify theoretically the expected population density of different states, we performed spectroscopy measurements at the plasma to estimate quantities needed to input in a dedicated simulation software. Before I discuss the results of spectroscopic measurements, I present some theoretical background to explain the methodology to quantify population density of states in plasma.

Discussion of the Results of Plasma Spectroscopy

In order to determine the plasma parameters, we did spectroscopic measurements on the plasma. The measurements were taken by randomising values of pressure and incident microwave power in the range of 0.2 mbar-1.2 mbar and 8 W-50 W respectively. Before taking measurements, the cavity was tuned to minimise the reflected power. At 50 W, the reflected power was only 1 W. Tuning the cavity avoids plausible errors in the data and maintains consistency in the results.

The two most intense transition lines found in the plasma are the Balmer- H_α (656 nm), H_β (486 nm). The line method can be applied to these two transitions to determine the population density distribution in the plasma. For every setting of pressure and incident power, five spectrums were recorded. H_α , H_β , H_α/H_β intensities were plotted with pressure and incident power. The standard deviations of intensities, ratio of intensities were also plotted (ref. fig. 4.3, 4.4, 4.5).

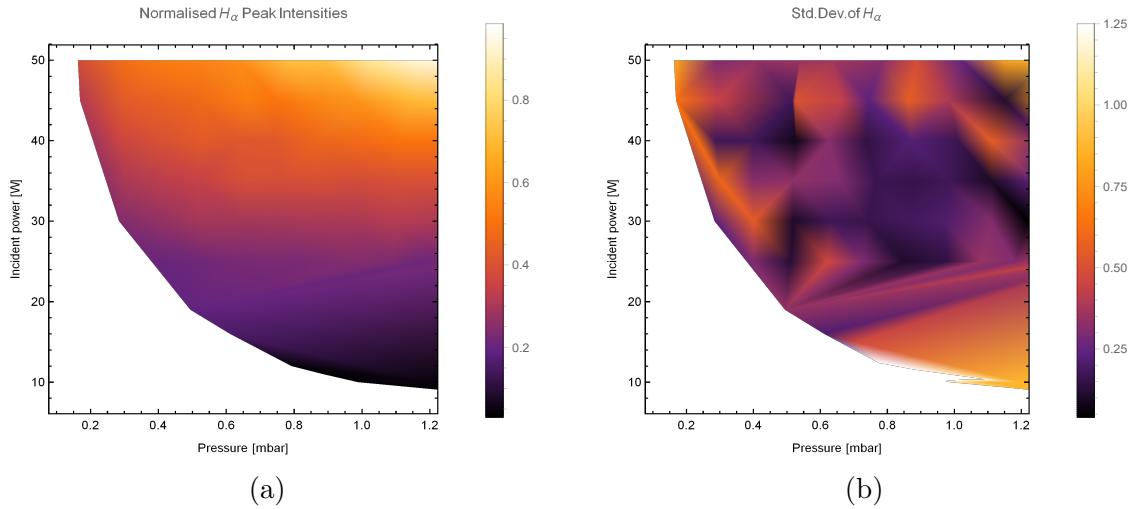


Figure 4.3: a) Normalised intensity plot of the Balmer- H_α line with varying H_2 gas pressure and incident microwave power. b) Standard deviation of data plotted in (a). A maximum deviation of $\sim 1.1\%$ is observed at low powers and high pressures.

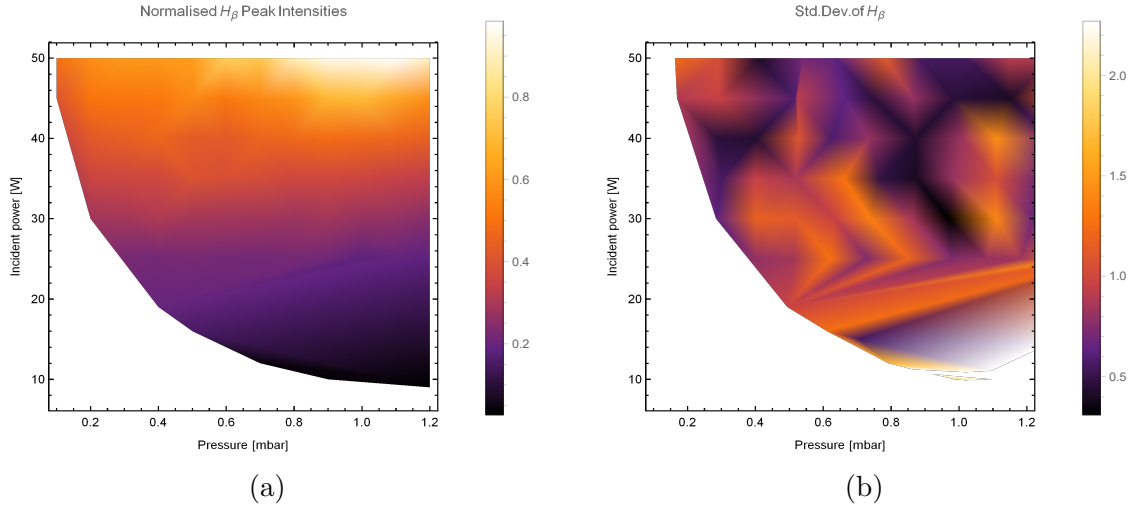


Figure 4.4: a) Normalised intensity plot of the Balmer- H_β line with varying H_2 gas pressure and incident microwave power. b) Standard deviation of data plotted in (a). A maximum deviation of $\sim 2\%$ is observed at low powers and high pressures.

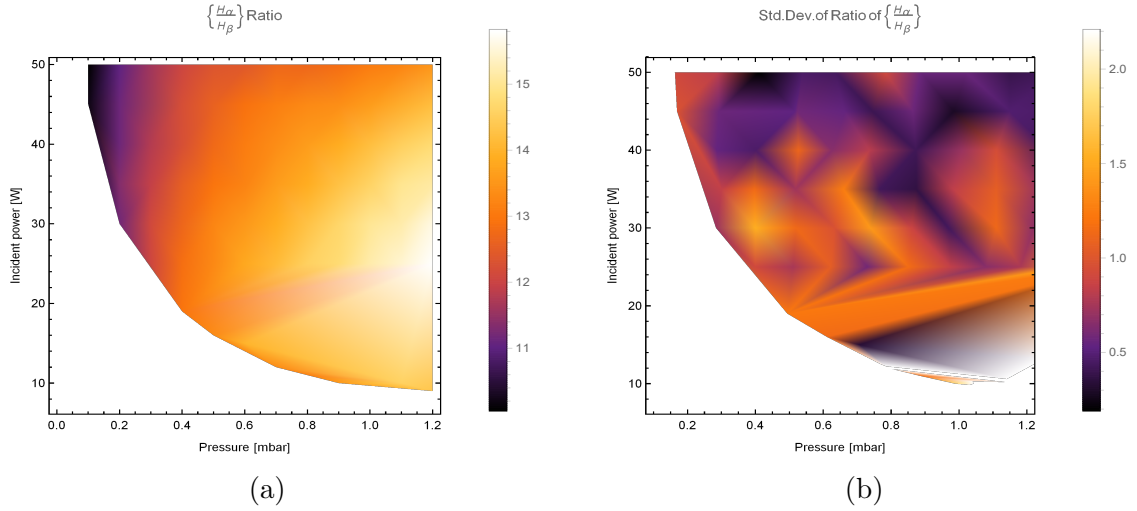


Figure 4.5: a) Plot of intensity ratio of the Balmer H_α/H_β lines with varying H_2 gas pressure and incident microwave power. The intensity ratios lie between 10-16. b) Shows the standard deviation of data plotted in (a). A maximum deviation of $\sim 1.8\%$ is observed at low powers and high pressures.

In the plots 4.3, 4.4, 4.5, it is observed that:

1. **For H_α, H_β :** The transition intensity is independent of pressure upto ~ 22 W, but later increases as the pressure is increased. The intensity always increases with respect

to power. The highest intensity is found at highest power and pressure. Maximum standard deviations are $\sim 1.1\%$ for H_α and $\sim 2\%$ for H_β . They occur at low powers ($< 14\text{ W}$). The standard deviation is the highest at low powers because the power is barely enough for the plasma to sustain, so it fluctuates and therefore is on the verge of being extinguished.

2. **For the ratio of (H_α/H_β):** Similar to the behaviour of H_α, H_β intensities, their ratio is independent of pressure upto $\sim 22\text{ W}$ and later increases as the pressure is increased. High values of the ratio are found in the intermediate power range $\sim 22 - 30\text{ W}$ and at high pressure $0.9\text{ mbar}-1.2\text{ mbar}$. The plot of standard deviation shows similar behaviour to those of the H_α, H_β . The maximum value is $\sim 1.8\%$ at low powers ($< 14\text{ W}$).

Population Models and Methodology to Quantify Population Density of States

There exist numerical codes that model the dynamics within the plasma. By matching particular line ratios obtained from those codes and comparing to the experimental data, one can estimate the plasma parameters.

There are mainly three kinds of population models²⁷:

1. **Corona models:** They are valid for plasmas with very low electron densities ($n_e \lesssim 10^{16}\text{ m}^{-3}$).
2. **Local Thermodynamic Equilibrium (LTE) models:** The LTE models are used when the electron density is very high ($n_e \gtrsim 10^{22}\text{ m}^{-3}$).
3. **Collisional Radiative models:** CR models are typically used for intermediate values of electron density ($\sim 10^{16} - 10^{22}\text{ m}^{-3}$).

The CR population models provide the broadest application range. Usually, we can apply these models to plasmas with a collision rate too high for corona models. Nonetheless, CR models intrinsically include the corona model, and for high collision rates, their results should converge to the population distributions predicted by the LTE models. A web application called "[Yacora on the Web](#)" provides access to collisional radiative models. One needs to type in a few parameters like the electron density and the plasma temperature to run the model. This application shows an excellent correspondence between measured and calculated emission intensities. Furthermore, the agreement between plasma parameters from other diagnostics and those derived using Yacora on the web from optical emission spectroscopy were also found to be very similar (ref.²⁸).

Typically the plasma we generate have densities of $\sim 10^{14} - 10^{15} \text{ m}^{-3}$ and temperature of a few eV. So, it is categorised as a recombining plasma. Since Yacora intrinsically considers corona models (valid for the n_e we have) and CR models that consider many more excitation channels than corona, we plan to use them to simulate the distribution of the populated states numerically. We plan to fit the results of the Yacora model to our spectroscopic data and find the parameters like n_e (electron density), T_e (electron temperature) and T_H (H atomic temperature). To determine these parameters, we use a method called the "**Line Ratio method**". The method is as follows:

1. First, by guessing plasma parameters the differential equation for population density (Eq.(1) from ref.²⁸) is solved, and the population coefficients are found at different values of n_e and T_e .
2. With population coefficients, the ratio of any two emission lines in terms of an "effective emission rate coefficient" (X) is calculated. X embeds information of the parameters n_e and T_e .
3. By comparing experimental line intensity ratios with simulated ones, we can alter n_e

and T_e until the simulations fit the experimental data.

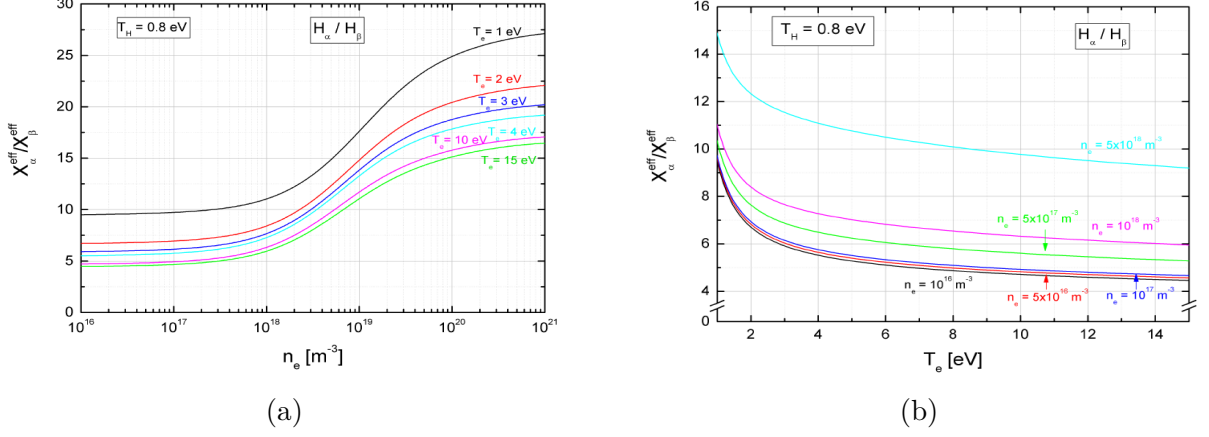


Figure 4.6: Produced with Yacora (adapted from ref.²⁹): Dependence of the ratio of effective emission rate coefficients on the a) electron density (n_e) and b) electron temperature (T_e), considering only the direct excitation channel. The Hydrogen temperature is constant ($T_H = 0.8$ eV). The $T_e = 1$ eV curve in fig. (a) can be approximately extrapolated in the $10^{14} - 10^{15} \text{ m}^{-3}$ region. In fig. (b), for low n_e values, the ratios are constricted in the range $8 \leq X_\alpha^{\text{eff}}/X_\beta^{\text{eff}} \leq 10$. This is a good indicator to predict the behaviour of the $n_e = 10^{14} - 10^{15} \text{ m}^{-3}$ curve.

Some results based on the line ratio method using Yacora have been reported in ref.²⁹ for recombining plasma. I analyse those results and compare them with the experimental results from our plasma spectroscopy.

The plots in fig. 4.6 do not contain the exact parameters that we expect our plasma to have *i.e.*, $n_e = 10^{14} - 10^{15} \text{ m}^{-3}$ and $T_e < 1$ eV, but some extrapolations can be made to predict the parameters. In fig. 4.6(a), the $T_e = 1$ eV curve stays approximately constant for $n_e = 10^{17} \text{ m}^{-3}$ and below. So, we can expect the simulated Balmer- H_α/H_β line ratios to be close to 10 (for $n_e = 10^{14} - 10^{15} \text{ m}^{-3}$), and indeed the ratios found in fig. 4.5 are close to 10 in the range of $10 \leq X_\alpha^{\text{eff}}/X_\beta^{\text{eff}} \leq 16$. In fig. 4.6(b), n_e curves converge in the lower T_e limit (≤ 1 eV). So for $n_e = 10^{14} - 10^{15} \text{ m}^{-3}$, we can expect similar convergence, very close to $8 \leq X_\alpha^{\text{eff}}/X_\beta^{\text{eff}} \leq 10$. It should be noted that the parameter held constant in both of the graphs is T_H (temperature of atomic H). So, a parametric scan of T_H near ~ 0.8 eV could

yield accurate values of the n_e and T_e . Then, using the line ratio method in reverse, we can evaluate the differential equation for population density and find the population of the $2s$.

4.2.2 Production of the 366 nm Beam

The main components of the laser setup (ref. fig. 4.9) are a commercial Nd:YAG pump laser (ref. fig. 4.7), a Ti:Sa cavity (based on the design from ref.³⁰. Ref. fig. 4.8), a seeding laser (ref. fig. 4.10) and a BBO frequency doubling crystal.

The Nd:YAG laser is a pulsed Class 4 laser. It is capable of producing 6 ns pulses with an average pulse energy of 20 mJ up to 20 Hz (we set it to 10 Hz). The 1064 nm beam it produces is internally frequency-doubled to 532 nm. At the exit of the laser is a half-wave plate that helps in optimising the intensity of the beam. A polarisation cube separates the parallel and perpendicular polarised components of the beam. Afterwards, the beam passes through a telescope that helps focus the beam. When the beam enters the cavity, two Brewster prisms help deviate the beam onto the Ti:Sa crystal (see fig. 4.8). These prisms produce total internal reflection and pass on the beam intensity like a quasi-achromatic mirror. The 532 nm beam passes through the Ti:Sa crystal and lases 732 nm light when the beam spot of the pump is correctly aligned on the crystal. The lasing occurs at an average pulse energy of ~ 120 mJ (of the YAG pump). The beam spot is about 2 mm in radius. So the energy density impinging on the Ti:Sa is ~ 2.5 J/cm². The Ti:Sa crystal has a threshold energy density of $\sim 8 - 10$ J/cm², which is well within the limit. A beam of 732 nm is injected into the cavity using a diode laser (see fig. 4.10) that seeds the lased beam. A coupler (semi-transparent mirror) couples the diode beam with the cavity. For seeding, the cavity has to be aligned and tuned, meaning that the resonant modes must be established corresponding to the wavelength of the diode laser. This is done by tweaking distances between cavity components. The buildup of resonant modes is extremely sensitive to minor adjustments.

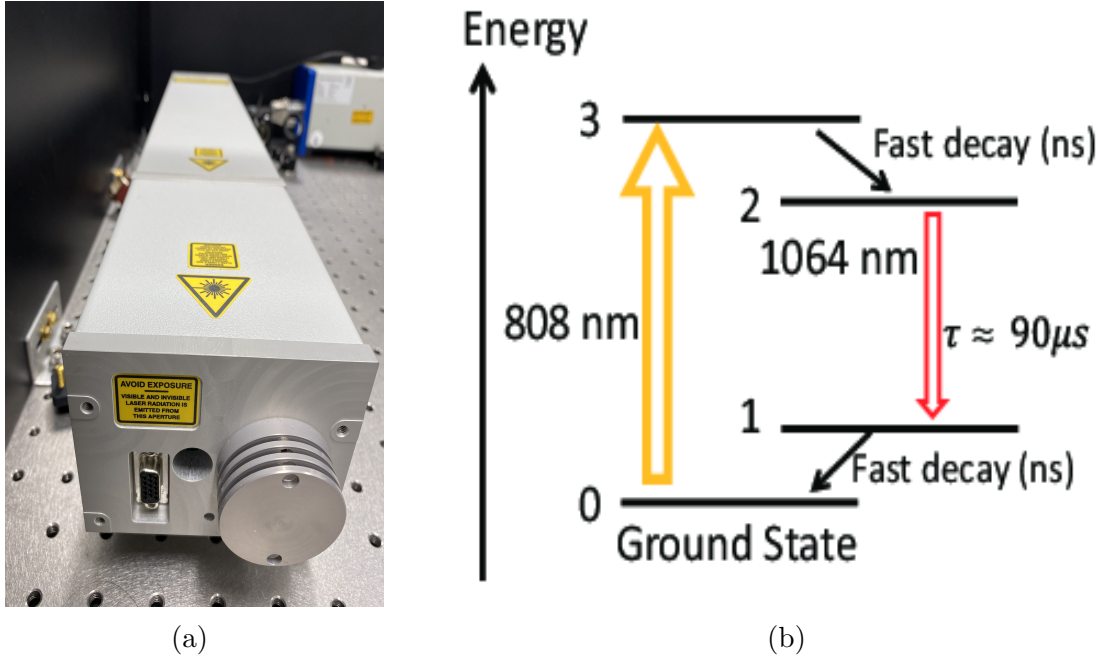


Figure 4.7: a) Picture of the Nd:YAG Pump Laser. b) Energy diagram of the lasing transition: A Krypton flashlamp optically pumps electrons to the highest level in a 4 level system. Spontaneous decay occurs to the $^4F_{3/2}$ metastable state. Lasing (at 1064 nm) occurs through the transition $^4F_{3/2} \rightarrow ^4I_{11/2}$ in Nd^{3+} ions. It is frequency doubled in the end to produce 532 nm beam.

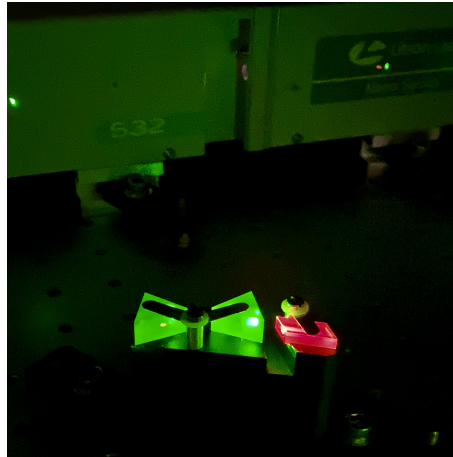


Figure 4.8: The green crystals are the Brewster prisms that help deviate the 532 nm pulsed beam and the 732 nm seeding beam and into the Ti:Sa crystal (in red). The Ti:Sa converts 532 nm beam into 732 nm. To seed this beam, cavity needs to be tuned.

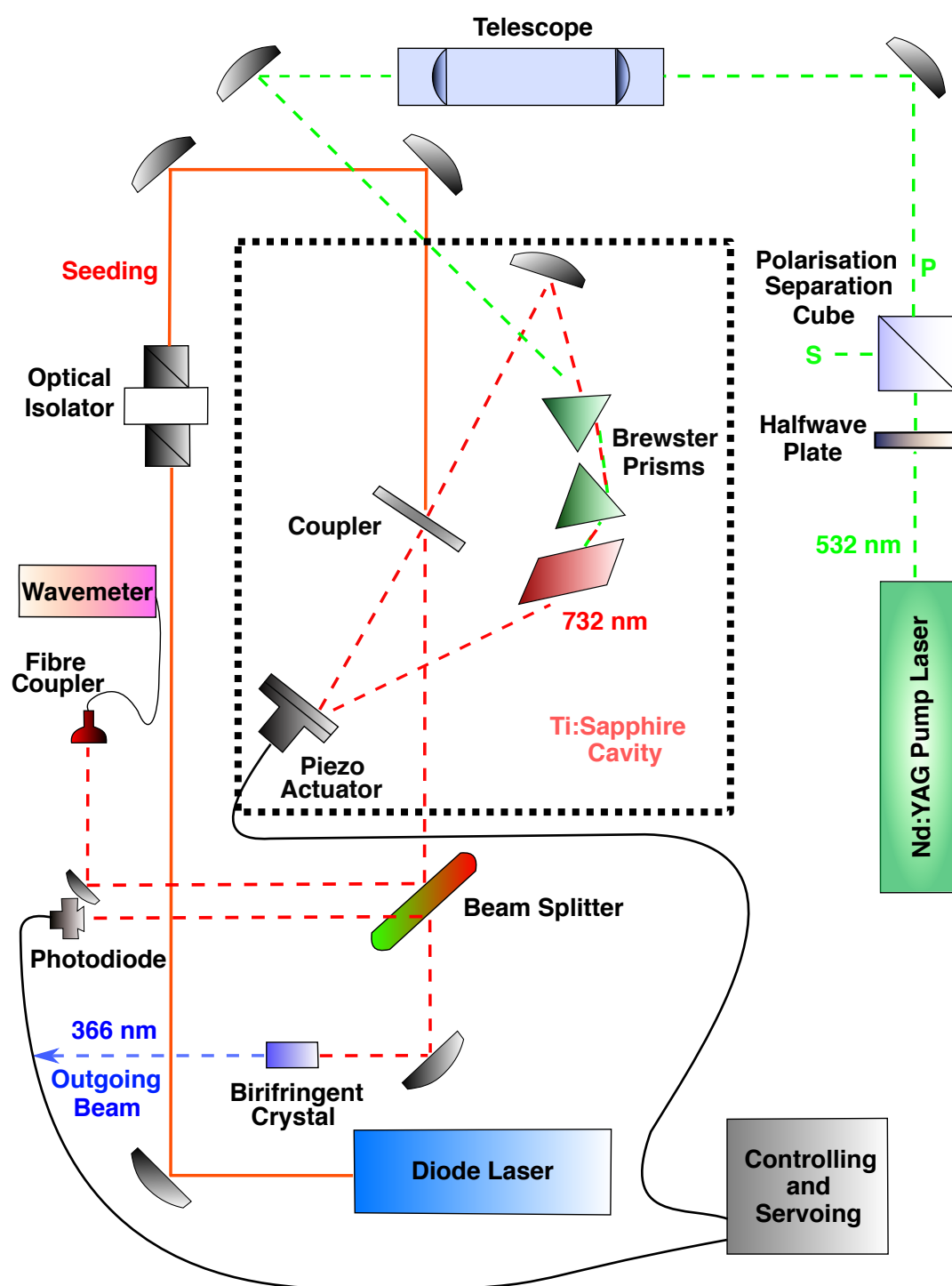
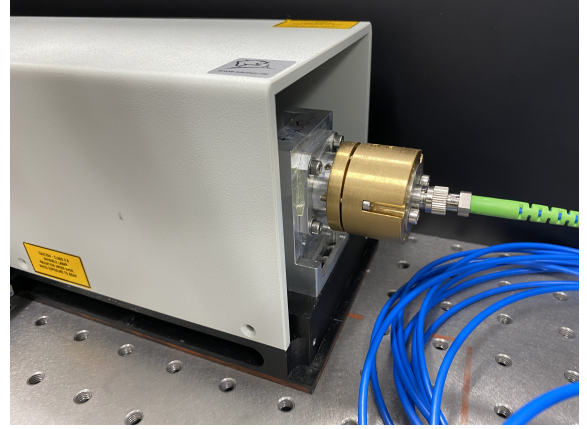


Figure 4.9: Schematic diagram of the laser setup



(a) Front view



(b) Side view

Figure 4.10: Picture of the seeding diode laser: It can be tuned in the range of 723-738nm. For our purpose, we set it to 732 nm. It is used to seed the red lasing beam in the Ti:Sapphire cavity. The diode laser is capable of supplying upto ~ 30 mW of power.

A photodiode sensor connected to an oscilloscope is used to observe the building resonant modes while the cavity is aligned. The modes can be seen building up in the form of periodic oscillations, and we aim to maximise the amplitude of this signal. A piezo actuator is attached to a mirror controlled by a PID system. It can be controlled with a computer program. The PID continuously adapts to the minor signal changes, which helps in maintaining resonance in the cavity. The wavelength of the diode laser has to be tuned, and tracked as it can change with environmental factors (like temperature). We track the wavelength by splitting the beam with a beam splitter and coupling one of the split beams into a fibre coupler connected to a High Finesse wavemeter. The other beam is directed towards a frequency-doubling BBO (Beta barium borate) crystal which converts the 732 nm beam to 366 nm. A transfer stage was built to pass the 366 nm beam into the beamline (see fig. 2.1). The excitation of H atoms is discussed in the next subsection.

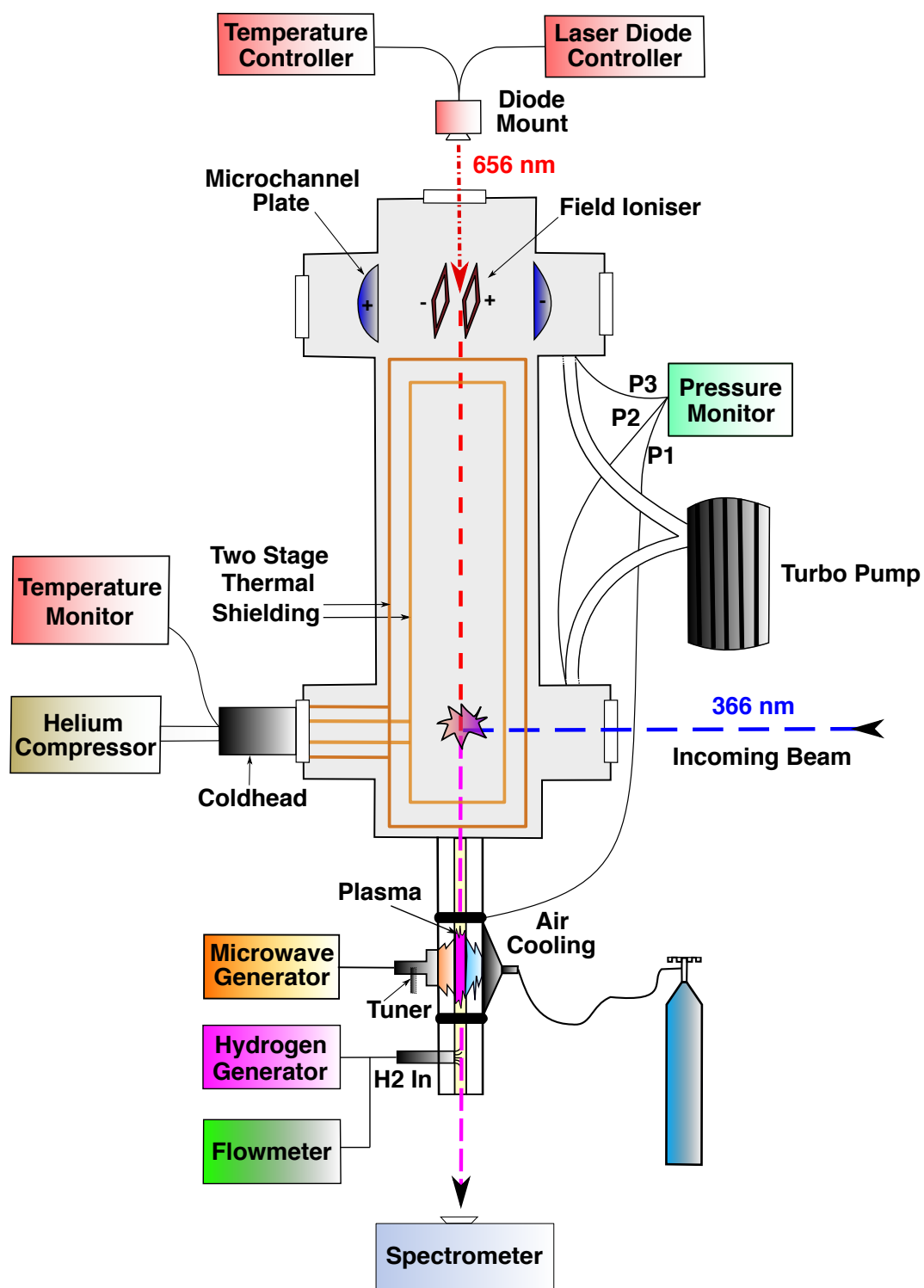


Figure 4.11: Schematic diagram of the beamline

4.2.3 Excitation of H and Detection of Rydberg States

The 366 nm beam enters the beamline through the transfer stage and perpendicularly crosses the path of H atoms coming out of the plasma. This is the region where the 2s will be excited to the $n=30$ circular state in presence of external $\mathbf{F}$ and $\mathbf{B}$ fields (to be implemented in the future).

The beamline is maintained at low vacuum pressures with two turbopumps attached to a backing pump that differentially pump out air from the excitation and the detection regions. The pressure in the detection region is about $\sim 10^{-8}$ mbar when the plasma source is off and can go to $\sim 10^{-5} - 10^{-6}$ mbar when on. Rydberg states are easily quenched or ionised by blackbody radiation. To prevent this, a two-staged copper thermal shield is installed that covers the area starting from where the H atoms enter the beamline until the beginning of the detection region. It is to shield blackbody photons of 300 K. A Helium compressor cools the inner copper shielding down to ~ 40 K.

H atoms travel about ~ 40 cm to reach the detection region where a Field-Ioniser(FI) and two Microchannel-Plates (MCPs) are placed (see fig. 4.12). The FI is made of two thin metallic frames with fine grids in the middle. One plate of the FI is connected to a high voltage source, and the other one is grounded. The FI produces an electric field potential that causes Rydberg H atoms to ionise. They fly in opposite directions to the two perpendicularly placed MCPs w.r.t. to the incoming H beam. These ionised particles then impinge on the MCPs. MCPs are charge amplifiers with many spatial channels. Depending on the magnitude and polarity of the high-voltage at the FI and on the microchannel plates (MCPs) located on each side of the beam, we can either detect e^- s or p^+ s impinging on the MCPs.

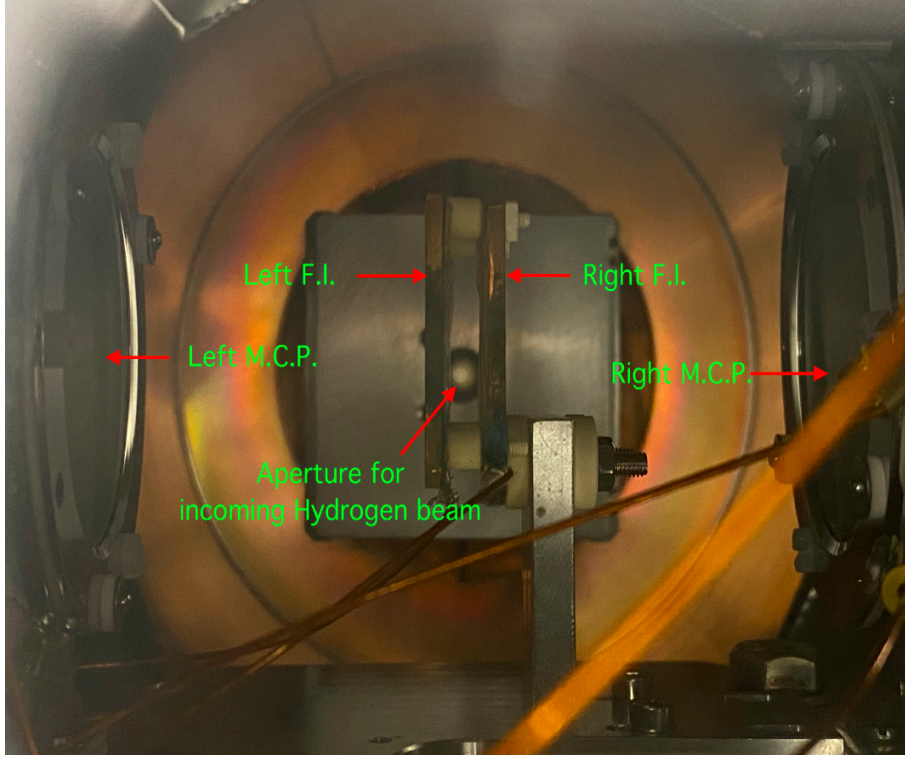


Figure 4.12: Picture of the detection region. In the middle: The excited Hydrogen beam comes through an aperture and the field ionisers ionise the atoms. On the sides: The particle count is detected by two Microchannel Plates on either sides of the chamber. For details on how MCPs work, see ref.³¹.

4.2.4 Doppler Spectroscopy with the Laser Diode

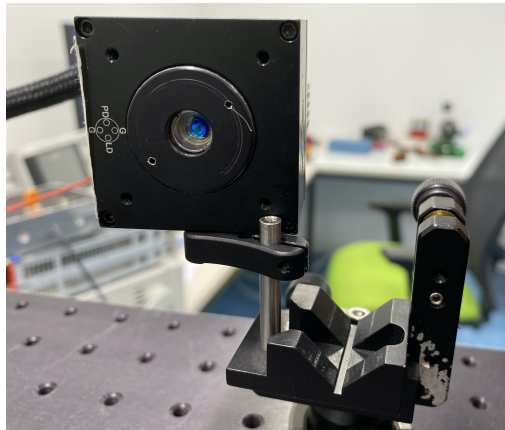
In order to characterize the hydrogen beam velocity, the group plans on performing Doppler spectroscopy of the $2s$ - $3p$ transition. Doppler spectroscopy measures the broadening (shift in frequency) of a transition line, in our case, of the $2s$ - $3p$ transition. The wavelength for $2s$ - $3p$ transition is 656.2793 nm for zero doppler shift. We plan to set our temperature-controlled laser diode at this frequency(see fig. 4.13).

The laser diode is be placed in front of the detection chamber as shown in the fig. 4.11. By measuring the Doppler shift in $2s \rightarrow 3p$ transition, the statistical distribution of the velocities is measured and hence the mean velocity. The expected velocity of the H atoms is of the order of $\sim 10^3\text{ m/s}$. When the $2s \rightarrow 3p$ transition is excited, just like any

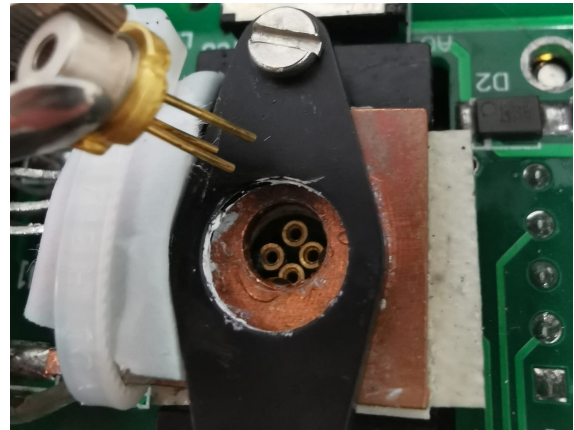
other excitation it spontaneously decays and emits radiation. This radiation can be detected by the MCPs. The number of $2s$ atoms in the detection chamber can be estimated via the measurement of the $3p \rightarrow 2s$ photons.

Precisely setting the wavelength of a temperature-controlled diode laser requires knowledge of the NTC thermistor in the mount. There exists a particular relation between the resistance/temperature applied and the wavelength that the laser diode produces. Through this relation, one can figure out the required resistance or temperature for the desired wavelength. It can be set on the temperature controller (see fig. 4.14). A PID system helps stabilise the resistance/temperature. We use a High Finesse wavemeter to temporally track the wavelength of the diode laser to calibrate it and track its stability. The laser controller is mainly used for controlling current applied on the laser diode which affects the intensity of the beam.

I contributed to stabilising the wavelength of the laser diode. At first, the wavelength could not be set to the desired temperature because the Peltier element had a cooling issue. It could not cool below a specific temperature. So we replaced the thermal paste. That did not work, so I decided to cool the mount externally. I simply resolved the problem by using a fan to cool the laser diode mount. After implementing this solution, the wavelength seemed stable only to the first decimal digit (in nm). The wavelength stability issue needs to be investigated further.



(a) Front View



(b) Internal View

Figure 4.13: a) Picture of the mount that contains the Laser Diode for Doppler spectroscopy. It can be controlled using a Laser controller and a Temperature controller. b) A Thermo-electric Cooler (TEC) is situated under the copper plate that works on the principle of the Peltier effect to control the temperature of the diode (in silver-gold colour). A Negative Temperature Coefficient (NTC) thermistor helps in setting and sensing the temperature of the diode. The wavelength of the diode can be tweaked around 656 nm by changing the temperature.

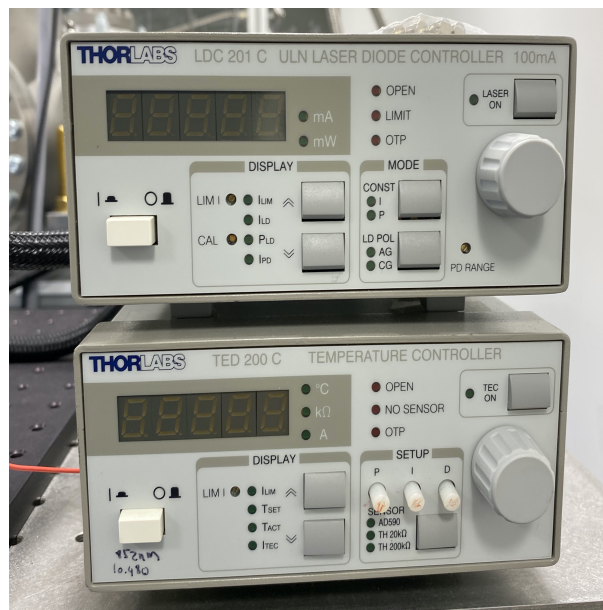


Figure 4.14: Picture of the Laser and Temperature controllers. Current acting on the diode can be set using the laser controller that directly affects the intensity of the beam. The temperature of the Peltier element can be controlled by changing the resistance of the NTC thermistor. A PID controller helps stabilise the temperature.

4.2.5 Data Acquisition with LabVIEW VI

The aim of programming a LabVIEW VI was to make data acquisition efficient by integrating all sensors and controlling them individually using a single interface.

The VI consists of a front panel and a block diagram. The front panel (see fig. 4.15) has several tabs corresponding to different sensors: temperature readout, pressure readout, microwave generator, Hydrogen flowmeter, spectrometer and the DAQ (data acquisition). Every sensor can be individually turned on or off using the start-stop buttons even when the VI is running. The block diagram contains several sub-VIs assigned to every sensor like the spectrometer sub-VI (see fig.4.16) for data logging of various parameters like pressures, temperature. The sub-VIs help in integrating specific features included in the instrument's driver. The sub-VIs can either be programmed using LabVIEW functions or sometimes they can be found in the LabVIEW drivers available on the instrument manufacturer's website. The main functions of the programmed LabVIEW VI are:

1. It can be used to continuously monitor pressures in the 3 regions (near the plasma source, in the excitation chamber, in the detection chamber) with a live waveform graph.
2. Similarly, it shows a live waveform graph of the temperature of the cooled thermal shielding.
3. We can control the incident power and limit the reflected power of the microwave source.
4. The customisations in the spectrometer sub-VI enable peak detection along with calculating peak ratios. However, a minimum intensity and a wavelength range have to be inputted for the peak ratio feature to work.

5. The most crucial feature of the VI is that the DAQ Sub-VI (see fig. 4.17) can integrate data from all sensors and save it in a single text file in a timely synced manner.

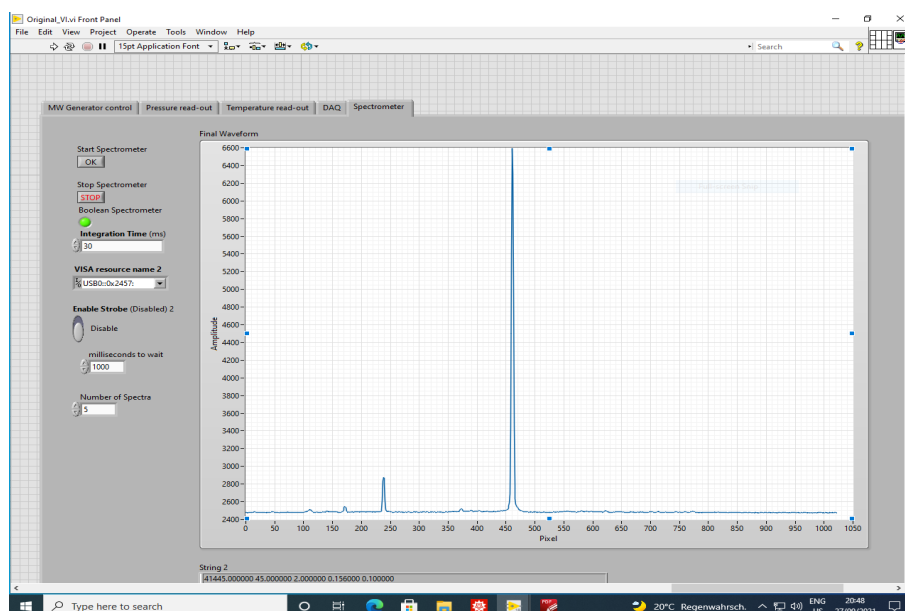


Figure 4.15: LabVIEW Front Panel

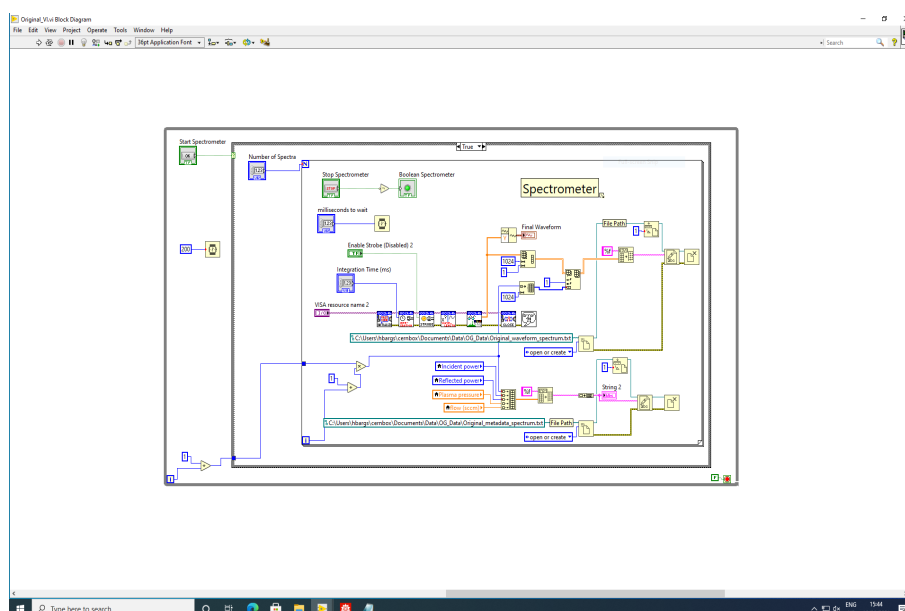


Figure 4.16: LabVIEW Spectrometer Sub-VI

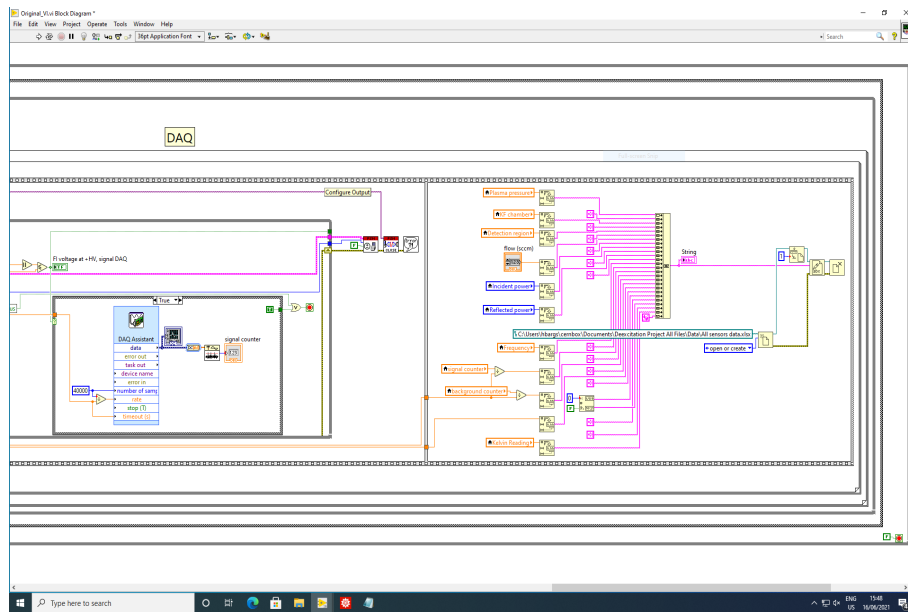


Figure 4.17: LabVIEW Data Acquisition Sub-VI

Chapter Five

Conclusion and Outlook

In this thesis, the theoretical and experimental developments of the proof-of-principle H excitation experiment were discussed.

In the theoretical part, first, the methodology to produce circular state atoms was presented. The atoms are laser-excited to low m states in the presence of external $\mathbf{F}$ and $\mathbf{B}$ fields, then the $\mathbf{F}$ field is turned off adiabatically holding the $\mathbf{B}$ field constant. The adiabaticity condition to turn off the $\mathbf{F}$ field was given by Eq. 3.16. Next, a numerical simulation was done that aimed to calculate optimal $\mathbf{F}$, $\mathbf{B}$ fields and their orientation to excite atoms to the $n=30$ circular state from the 2s state. It was found that if an electric field of $\mathbf{F} = 3000 - 5000$ V/m is turned off within $10\mu\text{s}$ within a distance of 5 cm along the beam path, then the adiabaticity criterion is satisfied while keeping the magnetic field of $\mathbf{B} = 20$ G constant. It was also estimated that the sub-states in the range $6 \leq l \leq 29$ would be detected at the expected velocity of the H atoms (a few 1000 m/s).

In the experimental part, the H plasma source was characterised by optical emission spectroscopy. The two transition lines, Balmer- H_α , H_β were studied. The H_α/H_β line ratio was plotted. The experimental line ratios were used to approximately extrapolate plasma parameters. It was done by comparing the line ratio plots produced using Yacora, a program

that uses collisional radiative model to calculate plasma parameters. The intensity ratios extrapolated from both the graphs were in close agreement to the ones that were found through the spectroscopic measurements. It was concluded that the parameter T_H has to be varied around 0.8 eV until the simulated line ratios fit with the measured ones. Once the parameters are found, the population density of states within the plasma can be quantified.

In the last chapter, the conceptual design of the experimental apparatus was explained through schematic diagrams. The production of a pulsed ~ 366 nm laser beam via a custom-built Ti:Sa laser pumped by a Nd:YAG was discussed. Further, the details of the envisioned Doppler spectroscopy of the $2s \rightarrow 3p$ to measure the velocity of H atoms and to detect 2s states with MCPs was explained. Finally, the features of a custom LabVIEW software built to control sensors and acquire time-synced data were described.

5.1 Outlook

The tasks that remain before the execution of the H excitation experiment are:

1. Characterise plasma by comparing Yacora simulations to the measured data of the plasma spectroscopy to evaluate its 2s H content. Note: In section 4.2.1, the plasma parameters were only approximately extrapolated based on the available Yacora data.
2. Perform Doppler spectroscopy on the $2s \rightarrow 3p$ transition to know H atoms' mean velocity and experimentally measure the 2s content via photon detection by the MCPs. If the 2s content is insufficient, other techniques have to be employed to produce 2s or 2p H.
3. Implement electric and magnetic fields into the apparatus and demonstrate circular state excitation.

We recall that the main goal of the H excitation experiment is to produce a source of highly excited atoms on which the stimulated deexcitation can be demonstrated. Many techniques to deexcite antihydrogen have already been studied (ref. [6,8](#)). If these techniques are proven feasible and efficient in practice, they would be implemented in the AE $\bar{g}$ IS and ASACUSA experiments. That would address the long-standing issue of antihydrogen deexcitation enabling beam experiments to produce useful beams of groundstate antihydrogen atoms.

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