

An ultracold beam of YbF molecules for measuring the electron's electric dipole moment

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Abstract

The observed matter-antimatter asymmetry of the universe remains unexplained and undiscovered sources of CP violation could yet prove to be its source. The electric dipole moment of leptons, strongly suppressed in the Standard Model, would be a source of CP violation and a sign of new physics. Recent measurements of the electron EDM have already pushed constraints on the energy range for new particles to be discovered beyond the reach of the LHC.

This work focuses on the progress towards an experiment to measure the electron EDM using YbF. A buffer gas-cooled molecular source generates a pulsed beam of YbF molecules travelling at 180 ms^{-1} . Employing two-dimensional laser cooling, the transverse temperature of the beam drops below $200 \mu\text{K}$, significantly increasing its brightness by over two orders of magnitude compared to the uncooled beam. On average, each cooled beam pulse contains approximately 2×10^5 ultracold molecules. A hexapole magnetic lens will guide the molecular beam towards the cooling lasers. Simulations predict that the lens will enhance beam brightness by at least five-fold. Combined with transverse laser cooling, this magnetic lens has the potential to yield a beam three orders of magnitude brighter. This enhancement will enable a measurement of the electron EDM, with a statistical uncertainty of $2.2 \times 10^{-31} e \text{ cm}$. This will represent an improvement of over an order of magnitude compared to the recent record established by JILA. Lastly, a new optical pumping scheme has been developed to transfer molecules from their laser cooling state to the absolute ground state of YbF, preparing for electron EDM measurement. This work lays the foundation for a high-precision measurement of the electron EDM using YbF.

Declaration

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Author's Contributions

My role was multifaceted throughout this work's research and development phases, entailing both individual undertakings and collaboration with a dedicated team. Central to our research was the groundbreaking achievement of two-dimensional laser cooling of YbF molecules. This milestone, which had not been reached previously, was achieved through the collective efforts of our team. I played a pivotal role in this endeavour, from building the experiment and making measurements to data analysis. Our collaborative efforts culminated in the publication of a paper that detailed our innovative laser cooling techniques, to which I contributed as a co-author [1].

In addition to these collaborative strides, I undertook an independent effort to design a magnetic lens for focussing the molecular beam into the cooling lasers. From its inception to its realisation, this project was an individual endeavour. While I did benefit from advisory inputs from colleagues, the design's conception, its subsequent construction, and the characterisation of the lens's properties were carried out by me. Our workshop's expertise was invaluable in manufacturing specific components essential for the lens.

Further expanding on the scope of my contributions, I was largely responsible for implementing a new optical pumping scheme. This innovative approach was designed to transfer molecules from their laser cooling state to the absolute ground state of YbF, setting the stage for the electron EDM measurement. While this venture was mainly an independent effort on my part, collaboration became essential during its optimisation phase.

My commitment to the project extended beyond these highlighted efforts. I delved into several other critical aspects of the experimental setup. One significant contribution was the creation of custom software that enabled the integration of the experiment hardware with the computer, automating various facets of our research process and enhancing efficiency. Alongside this software development, I significantly refined the buffer gas molecular source, ensuring a consistent and reliable YbF molecular beam. Furthermore, my involvement with the assembly and troubleshooting of numerous laser optics circuits was vital in our efforts to maintain reliable laser cooling of the molecular beam.

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Chapter 1

Introduction

In the realm of fundamental physics, the pursuit of knowledge extends beyond the boundaries of our current understanding as scientists strive to unravel the mysteries underlying the mechanics of nature. Among the phenomena that captivate physicists, high-precision measurements of the electron's electric dipole moment (eEDM) stand out as a highly anticipated research area. This chapter presents an overview of the eEDM, the motivations behind its study, the history of its measurement, and the advancements made in the current generation of eEDM experiments, particularly using ytterbium monofluoride (YbF).

In section 1.1, I describe the electron electric dipole moment and delve into the motivations that drive physicists to pursue high-precision measurements. This exploration includes the implications such measurements hold for fundamental symmetries of nature, the Standard Model, the quest to understand matter-antimatter asymmetry, and the search for new physics. Section 1.2 provides an overview of the historical eEDM measurements. Through this review, I discuss the challenges encountered throughout the years and the notable progress achieved in this field. In section 1.3, I present the general methodology employed in eEDM measurements using atomic and molecular beams. This section outlines the fundamental techniques and principles involved in such measurements. Finally, in section 1.4, I focus on the specific approach of measuring the eEDM using a YbF molecular beam. I describe the experimental setup and techniques employed, highlighting the advancements made in this thesis. I also explore the limitations of this method.

1.1 The electron electric dipole moment

Electric charges can be either positive or negative; when paired, they create a dipole. The electric dipole moment (EDM) quantifies the extent of their separation. Specifically, if you have two charges of opposite signs, each with a magnitude of q , and they are separated by a distance $\vec{r}$, their EDM is given by,

$$\vec{d} = q\vec{r}. \quad (1.1)$$

The axis connecting the two charges defines the dipole moment's direction. As an illustration, certain molecules exhibit a difference in electronegativity between their atoms. This discrepancy can induce charge separation, generating a dipole moment that aligns with the axis between the atomic nuclei.

The concept of an electron possessing an electric dipole moment challenges our intuition when considering its depiction in the Standard Model of particle physics as a point-like entity devoid of internal structure. This portrayal makes the classical idea of a dipole, relying on spatial charge separation, seem inapplicable. However, envisioning the EDM as an intrinsic property of electrons can be aided by an analogy to a distorted sphere. Just as a sphere, when uniformly charged, represents a perfect monopole charge distribution, a distorted sphere symbolises an asymmetry in this distribution. This asymmetry in the electron's charge distribution, akin to the bulges on a distorted sphere, affects how the electron interacts with an external electric field. The electron's EDM, therefore, isn't a physical structure per se but an intrinsic property that dictates its interaction with an electric field and, from the Wigner-Eckart theorem, this intrinsic asymmetry is not arbitrary but aligned or anti-aligned with the electron's spin [2, 3].

The pursuit of measuring the electron's electric dipole moment with heightened precision is propelled by the fundamental symmetries that underpin the laws of physics: charge (C), parity (P), and time-reversal (T) [1, 4–10]. These symmetries and their violations form the bedrock of our

understanding of particle interactions and the overarching structure of the Standard Model (SM) [11–13]. The discovery of a non-zero eEDM would be a clear indicator of new physics beyond the SM, particularly concerning CP violation, which is deeply intertwined with the matter-antimatter asymmetry observed in the universe [12, 14].

The Hamiltonian for a particle with a magnetic moment μ and an electric dipole moment d , in the presence of external magnetic ($\vec{B}$) and electric ($\vec{E}$) fields, can be expressed as:

$$\hat{H} = -\mu \hat{s} \cdot \vec{B} - d \hat{s} \cdot \vec{E}, \quad (1.2)$$

where $\hat{s}$ denotes the unit vector along the particle's spin $\vec{s}$.

Under a parity transformation, which inverts spatial coordinates, the behaviour of the Hamiltonian reveals symmetry violation. The magnetic field term, which involves the magnetic field $\vec{B}$ and the spin $\vec{s}$, is unchanged due to their pseudovector properties. In contrast, the electric field term, involving the electric dipole moment d and the electric field $\vec{E}$, would change sign. This asymmetry indicates that a non-zero value for the electron's electric dipole moment, d_e , would manifest as a parity symmetry violation [12].

Time reversal symmetry provides further insights. This transformation reverses the direction of the magnetic field and the spin but not the electric field. A non-zero d_e would, therefore, indicate a violation of time reversal symmetry, which, assuming the inviolability of the combined CPT (charge, parity, and time) symmetry, also implies CP violation [15]. CP violation is integral to the Standard Model, and its presence through an electron EDM could suggest new physics beyond what is currently understood. These theoretical underpinnings, rooted in the CPT symmetry—a foundational principle of quantum field theories—highlight the potential for groundbreaking discoveries through EDM measurements [15].

The potential discovery of a non-zero electron electric dipole moment exceeding the extremely small prediction of $d_e \leq 10^{-38}$ e cm within the framework of the Standard Model would provide

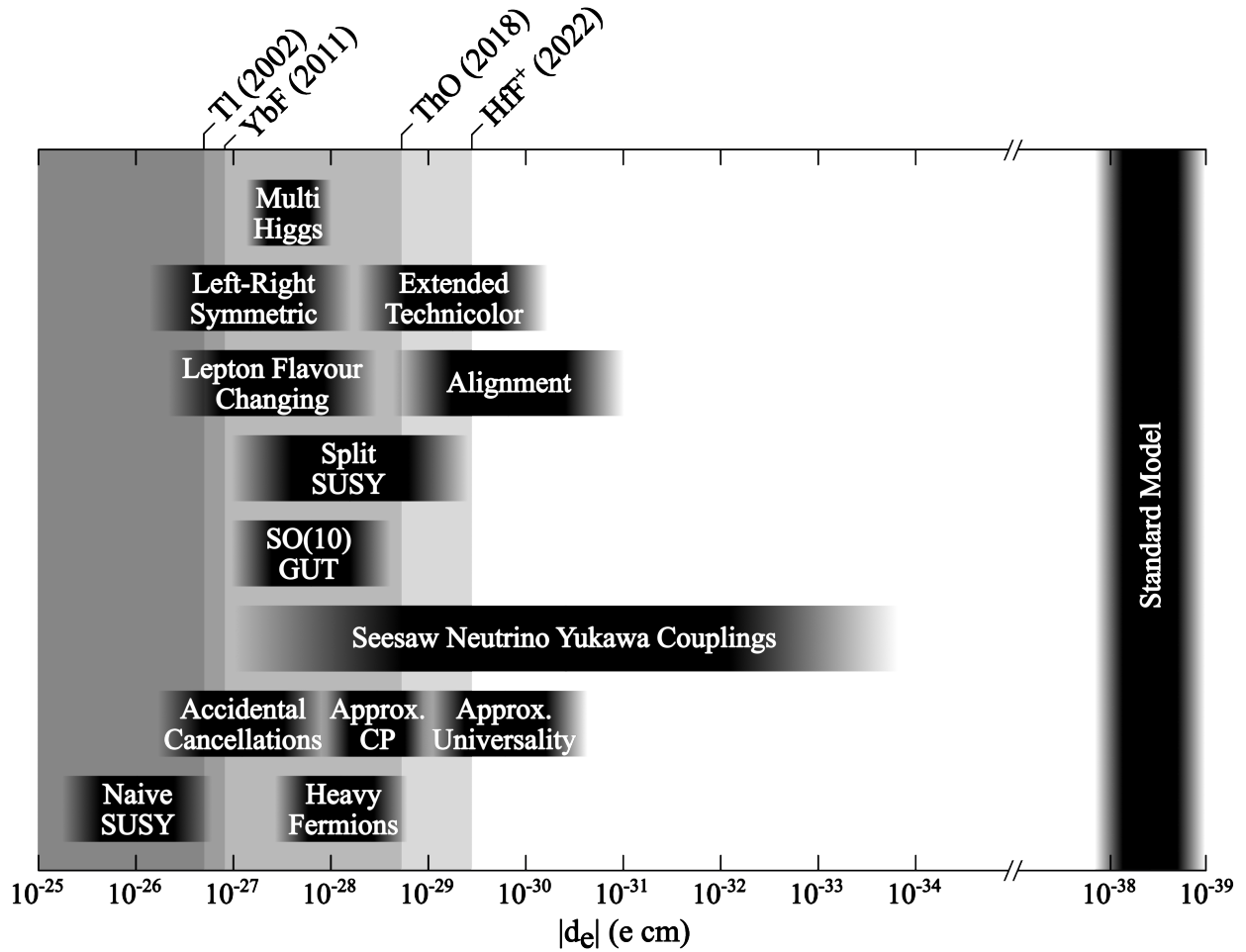


Figure 1.1: Comparative predictions and experimental constraints on the electron electric dipole moment (d_e). The minuscule prediction of d_e by the Standard Model is depicted alongside notably larger estimates from various Beyond Standard Model (BSM) theories. These BSM predictions are compared against the upper limits derived from experimental data, which are represented by the shaded grey regions. Each experiment is identified by the type of species utilised and is accompanied by the corresponding year of the measurement, with full details provided in Table 1.1. This figure refines and incorporates updates to the original illustration by Elizabeth Petrik (2014) [17].

compelling evidence for physics beyond the Standard Model (BSM) [12, 14]. This holds great significance due to the crucial role of CP violation in explaining the observed imbalance between matter and antimatter in the Universe [14]. By pushing the boundaries of precision in eEDM measurements, we can impose constraints on the potential energy scales associated with new fundamental particles. Notably, the recent measurement of d_e by the Cornell Group has placed constraints on the energies of new BSM particles, exceeding those reachable by the LHC [16].

Figure 1.1 shows the critical juncture at which current experimental results challenge many BSM theories. The figure's black boxes represent the predicted ranges for the magnitude of the elec-

tron electric dipole moment according to these theories. The grey areas indicate the upper limits established by the most recent and stringent experiments, as detailed in Table 1.1. Historically, the measurement of d_e has been an essential tool in exploring new physics, offering insights that could confirm or dismiss extensions to the Standard Model. Up to this point, many meticulous experimental studies have successfully imposed more restrictive boundaries on d_e , progressively ruling out large portions of the parameter space that many BSM theories predict.

Significantly, this includes a range of supersymmetry (SUSY) models, historically favoured for their elegant solutions to various Standard Model problems [18]. Naive SUSY models [19, 20], and models such as Accidental Cancellations, which rely on fine-tuned cancellations between different contributions to d_e , have seen their parameter spaces severely curtailed by experimental evidence [21]. Similarly, the stringent limits have also disfavored certain incarnations of Split SUSY and SO(10) Grand Unified Theory (GUT) models, as well as multiple Higgs, Left-Right Symmetric, and Lepton-Flavor changing models, which predict larger electron EDMs than what has been observed [22–25].

In contrast, the Seesaw Neutrino Yukawa Couplings model remains a viable theoretical space. This model offers a broad prediction for the electron EDM, which remains largely unchallenged by current measurements [26, 27]. The resilience of this model underscores the complexity of neutrino physics and the potential insights that might be gleaned from a measured electron EDM within its predicted range. Additionally, current experimental limits have not yet encroached significantly upon the predictions of Approximate Universality in the SUSY framework or Alignment models [28, 29]. These models predict smaller dipole moments not currently excluded by experimental data.

The work detailed in this thesis encompasses an experiment conceived to measure d_e with unparalleled precision. While the primary goal is not to target any specific BSM theory, the forecasted sensitivity of this experiment—provided in the numerical results presented later in chapters 4 and 5—indicates that it has the potential to seriously contest the parameter spaces of Approximate Universality and Alignment models within the supersymmetry framework.

1.2 Measurements of the electron EDM

In recent decades, a remarkable surge of research activity has been dedicated to measuring the electron's electric dipole moment. Numerous independent research groups have been racing to achieve increasingly precise measurements. The progress in this field is highlighted by a variety of methods and species used for these measurements, as shown in Table 1.1 [4, 5, 10, 16, 30, 31]. These diverse approaches demonstrate the fascinating advancements in controlled quantum dynamics within the field.

At their core, the techniques used in all of these experiments are based on a fundamental concept. The interaction of the eEDM with an applied electric field shifts the energies of the magnetic sub-levels in exactly the same way as the Zeeman splitting in a magnetic field. This can also be viewed in terms of the spin precession of the electron when polarised orthogonal to the applied field: a magnetic field causes the spin to precess, and an electric field increases or decreases that spin-precession rate depending on the relative orientation of $\vec{E}$ and $\vec{B}$. By preparing the atom or molecule in a superposition state involving non-zero magnetic sublevels, the precise measurement of the spin precession rate can be achieved through interferometric techniques. The discrepancy in the precession rates upon reversing the electric field can then unveil the value of d_e .

Electron EDM measurements never use bare electrons. The experiments are always done with atoms or molecules. The applied electric field polarises the atom or molecule, and it is necessary to calculate the total field seen by the electrons, including the internal polarisation field. At first glance, it seems as though the system must polarise in such a way as to make a net field of zero at every electron, for if it were not zero, the electron would be accelerated by the field and leave the system. This is true in the non-relativistic limit but not true more generally. In a highly relativistic atom, the total electric field is larger than the applied field. The discovery that a heavy atom actually enhances the eEDM interaction was a major stimulus to the field [32–35]. Because molecules are much more polarisable than atoms, they provide even greater enhancement factors and those large enhancements can be reached with modest applied electric fields. For this reason, most eEDM experiments now use molecules. The enhancement effect is typically captured by introducing an effective electric field E_{eff} . To give an example, a YbF molecule in an applied electric

field of 10 kV/cm has $E_{\text{eff}} \approx 14$ GV/cm, which is a very remarkable enhancement [3, 33].

Year	Group	Method	Species	Upper limit on d_e (e cm)
2002	Berkley	Atomic beam	Tl	1.6×10^{-27} [30]
2011	Imperial	Molecular beam	YbF	10.5×10^{-28} [4]
2014	ACME	Molecular beam	ThO	8.7×10^{-29} [10]
2017	JILA	Trapped molecular ions	HfF ⁺	1.3×10^{-28} [31]
2018	ACME	Molecular beam	ThO	1.1×10^{-29} [5]
2022	JILA	Trapped molecular ions	HfF ⁺	4.1×10^{-30} [16]

Table 1.1: Compilation of electron EDM measurements, all of which are consistent with zero. These measurements have been conducted using various experimental methods and different particle species, providing complementary measurements. Notably, over the course of two decades, the upper bound on d_e , determined with a 90% confidence level, has experienced a remarkable reduction by nearly three orders of magnitude.

In 2002, a measurement using an atomic beam of thallium atoms provided insight into the eEDM [30]. The measurement yielded a value of $d_e = (6.9 \pm 7.4) \times 10^{-28}$ e cm, with an upper bound of $|d_e| < 1.6 \times 10^{-27}$ e cm. To achieve this, the thallium beam was prepared in a superposition state of $m_F = \pm 1$ Zeeman sublevels and subsequent spin precession measurements were conducted in the presence of a strong electric field with a magnitude of $|\vec{E}| \approx 123$ kV/cm. This experimental approach is commonly employed in various eEDM measurements.

An interesting aspect of the study was utilising a second atomic beam consisting of sodium atoms. The sodium beam ran parallel to the thallium beam within the same experimental setup but with a slight separation of a few centimetres. While the sodium atoms served as comagnetometers, they were insensitive to the eEDM. This ingenious arrangement helped mitigate common-mode noise and systematic effects, providing a means to control the experiment's numerous magnetic noise sources [30].

Although thallium appears to be the only atomic species in table 1.1, other atomic species have also been considered and utilised in eEDM measurements. Caesium atoms, for instance, have been extensively employed in earlier studies due to the wealth of knowledge and controlled experimental techniques associated with this element [15, 36–38]. Similarly, francium has gained attention as an attractive candidate owing to its large enhancement factor [15, 32]. Current efforts are underway to create ultracold francium atoms within a magneto-optical trap, further advancing the exploration of eEDM [39, 40].

In 2011, a groundbreaking experiment using ytterbium monofluoride (YbF) molecules made a significant leap forward in precision molecular measurements [4]. The experiment, which served as a precursor to the research outlined in this thesis, employed a pulsed supersonic beam of YbF molecules cooled to about 4 K [2, 3, 41]. The measured value, $d_e = (-2.4 \pm 5.7_{\text{stat}} \pm 1.5_{\text{syst}}) \times 10^{-28}$ ecm, was consistent with zero but placed an upper limit of $|d_e| < 10.5 \times 10^{-28}$ ecm on the magnitude of the eEDM [4]. Similar to previous experiments, this study employed spin precession measurements to probe the eEDM.

The third and fifth rows of Table 1.1 highlight the remarkable achievements of the ACME collaboration. Their measurements of $d_e = (-2.1 \pm 3.7_{\text{stat}} \pm 2.5_{\text{syst}}) \times 10^{-29}$ ecm and $d_e = (4.3 \pm 3.1_{\text{stat}} \pm 2.6_{\text{syst}}) \times 10^{-30}$ ecm set eEDM precision records [5, 10]. Their experimental approach is similar to the YbF experiments conducted at Imperial College London [42]. The ACME collaboration generates a beam of cold ThO molecules using a pulsed cryogenic buffer gas beam source, similar to the design discussed in chapter 3 [43]. A spin precession measurement is then made using this beam. A significant achievement of ACME was the development of a stimulated Raman adiabatic passage (STIRAP) method for molecular spin state preparation. This innovative technique resulted in a remarkable increase in the number of molecules available for their eEDM measurement. In fact, their advancement led to a more than an order of magnitude amplification of the number of molecules in their measurement state [44]. Consequently, this substantial increase significantly reduced the statistical uncertainty of their second measurement compared to their initial measurement [5]. It is worth mentioning that we intend to employ this technique in the upcoming YbF experiment presented in this thesis.

A common objective in all the aforementioned experiments has been to enhance the number of particles under interrogation and extend the interaction time in spin precession measurements. This pursuit is driven by the aim of reducing the statistical uncertainty. However, the two HfF⁺ experiments conducted by the Cornell Group, as presented in Table 1.1, adopt a different approach [16, 31]. While larger molecule populations remain desirable, the Cornell Group prioritises significantly longer interrogation times by ionising their HfF molecules and confining them in a radiofrequency (rf) trap. This approach starkly contrasts the molecular beam experiments mentioned

earlier, which typically operate with interrogation times on the millisecond scale. In contrast, the Cornell Group can extend their molecular ion interrogation duration up to an impressive three seconds [5, 16].

To facilitate both the trapping and orientation of the molecular ions, the Cornell Group employs the rotation of the bias electric field at a frequency of $2\pi \times 375$ kHz [16]. While the notion of conducting high-precision measurements within the potentially noisy environment of an rf trap may seem counterintuitive, the results speak for themselves. In 2017, they achieved an eEDM measurement of $d_e = (0.9 \pm 7.7_{\text{stat}} \pm 1.7_{\text{syst}}) \times 10^{-29}$ e cm, followed by a subsequent measurement in 2022 with $d_e = (-1.3 \pm 2.0_{\text{stat}} \pm 0.6_{\text{syst}}) \times 10^{-30}$ e cm, which stands as the most precise eEDM measurement to date [16, 31, 45].

The Cornell Group is currently engaged in ongoing research exploring using ThF^+ molecular ions as a potential candidate for eEDM measurements, following their successful investigations with HfF^+ . ThF^+ exhibits promising characteristics as a competitive candidate, thanks to its effective electric field reaching up to 35 GV cm^{-1} [46]. Recent spectroscopic measurements and comparisons with theoretical calculations have even suggested the possibility of trapping ThF^+ ions for an impressive duration of up to 20 s [47]. The prospects of utilising ThF^+ ions in eEDM measurements are highly captivating, and if successful, they can revolutionise the precision of eEDM measurements beyond anticipated limits.

1.3 Measuring the electron EDM using atomic or molecular beams

Figure 1.2 depicts the general methodology for measuring the electric dipole moment (eEDM) using a beam of atoms or molecules. The process involves several steps: state preparation, spin alignment, interferometric interaction, and phase detection.

The atoms or molecules are initially generated from an atomic or molecular source. Specific state preparation procedures are employed as they travel down the beamline, tailored to the source type and the atomic/molecular species. These steps may include optical pumping to enhance

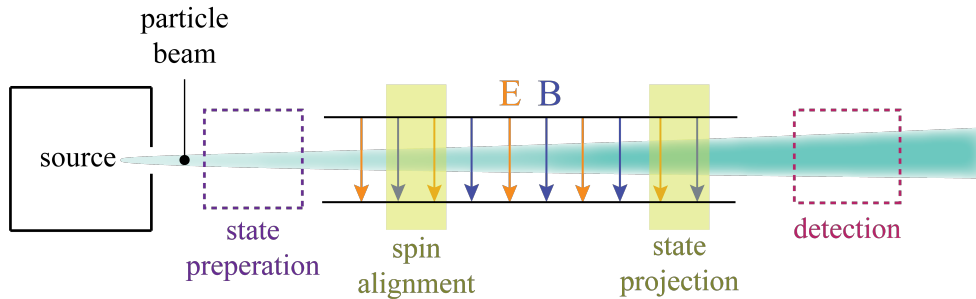


Figure 1.2: Illustration of the general methodology employed in electron electric dipole moment measurements utilising atomic or molecular beams.

the population of atoms or molecules in the desired eEDM measurement state, along with other preparatory actions.

Subsequently, the experiment follows a Ramsey-type interferometer setup. The electron's spin is first polarised by, for example, applying a π or $\pi/2$ -pulse. The beam then propagates through the interaction region. Within the interaction region, spin precession occurs at a rate that depends on the magnitudes and relative orientations of the applied fields.

After the interaction region, the spin precession angle is measured by mapping it onto a population difference between two quantum states. The populations are determined by counting the number of particles in each state, for example, by laser-induced fluorescence detection.

1.4 Measuring the electron EDM using a YbF molecular beam

The 2011 Imperial College measurement employed an apparatus consisting of a pulsed supersonic beam source, which generated a fast YbF molecular beam with a velocity of approximately 600 ms^{-1} [4]. The molecular beam propagated through a vacuum beamline. The initial step involved optically pumping the molecules into the absolute ground state $X^2\Sigma^+(\nu = 0, N = 0, F = 0)$, where ν , N , and F denote the vibrational, rotational, and total angular momentum quantum numbers, respectively (see section 2.1 for more details on the YbF molecular states).

Subsequently, the eEDM measurement was conducted. Initially, the molecules in the state $|F, m_F\rangle = |0, 0\rangle$ were prepared in a superposition state of Zeeman sub-levels, specifically $(|1, +1\rangle - |1, -1\rangle) / \sqrt{2}$, by applying a magnetic radiofrequency $\pi/2$ -pulse. This manipulation aligned the electron spin perpendicular to the applied magnetic field. The molecules then travelled for a duration of τ in the presence of uniform magnetic and electric fields, causing them to precess at a frequency ω and accumulate a phase $\phi = \tau\omega = \tau\Delta E/\hbar$. Here, $\Delta E = \mu B - d_e E_{\text{eff}}$ represents the energy shift induced by the applied fields, where μ is the magnetic dipole moment, B is the magnetic field strength, d_e is the electric dipole moment, and E_{eff} is the effective electric field.

The state evolves to $(e^{i\phi}|1, +1\rangle - e^{-i\phi}|1, -1\rangle) / \sqrt{2}$. At the end of the interaction region, ϕ was measured by projecting the superposition state back onto the $|0, 0\rangle$ state and independently determining the populations of the $F = 0$ and $F = 1$ states. These populations, which are proportional to $\cos^2(\phi)$ and $\sin^2(\phi)$, respectively, reveal the phase change. The Ramsey fringes could be observed by varying the field strengths during a series of measurements. However, the subsequent steps involved in precisely determining the eEDM are complex and go beyond the scope of this discussion. I recommend referring to Christopher Ho's informative thesis [3] for a more detailed understanding.

In an eEDM experiment constrained by quantum projection noise, the statistical uncertainty, commonly referred to as the "shot noise limit," exhibits an inverse relationship with the interaction time τ and is inversely proportional to the square root of the number of detected molecules $\sqrt{N_{\text{det}}}$ [3, 5, 48]. The fast velocity of the molecular beam utilised in the 2011 measurement imposed limitations on the interaction time between the beam and the fields. Conversely, a slower molecular beam confronts the challenge of thermal expansion, which restricts the number of detectable molecules.

To address these challenges, significant advancements have been made, as presented in this thesis. The implementation of a buffer gas-cooled molecular source yields a slower molecular beam, consequently extending the interaction time. Additionally, the application of transverse laser cooling effectively mitigates the issue of thermal expansion in the beam. These developments collectively enable the utilisation of a longer interaction region, further enhancing the interaction time avail-

able for the measurement.

Chapter 2

Theoretical foundations of the experiment

In this chapter, I delve into the theoretical foundations that form the basis of our experiment. It begins with an exploration of the molecular structure of the YbF molecule in section 2.1. Following that, I describe the interactions between YbF and external electric and magnetic fields, as outlined in section 2.2. Lastly, I explore the essential concepts of laser cooling in section 2.3. This section begins with a concise historical overview of laser cooling and explains the key light-matter interactions necessary for understanding laser cooling. Subsequently, I delve into the topic of Doppler laser cooling and conclude with a discussion of several sub-Doppler laser cooling techniques.

2.1 Quantum states of the ytterbium monofluoride molecule

This section is dedicated to exploring the molecular structure of ^{174}YbF . Ytterbium has several stable isotopes that occur naturally, but this work solely concerns the ^{174}Yb isotope. Therefore, when referring to YbF, it specifically denotes the ^{174}YbF isotopologue unless explicitly stated otherwise.

YbF is a diatomic molecule composed of one ytterbium atom and one fluorine atom. Ytterbium, a metallic element with atomic number 70 and electron configuration $[\text{Xe}] 4f^{14}6s^2$, is relatively electropositive. Fluorine, a halogen with atomic number 9 and electron configuration $[\text{He}] 2s^22p^5$, is the most electronegative element in the periodic table. When these two elements react, ytterbium

readily donates one of its 6s electrons to the 2p orbital of the fluorine atom, forming a monovalent Yb cation and an F anion, with an ionic bond between them, Yb^+F^- . The remaining valence electron of Yb is the 6s electron, which remains localised in the Yb^+ end of the molecule [49].

In the rest of this section, various aspects of the states in YbF are discussed, starting with the YbF electronic states. Following that, the rotational and vibrational states, which depend on the nuclear motion, are examined. Additionally, the interactions between YbF and electric and magnetic fields are explored. Finally, a discussion on transitions between states in YbF is presented.

2.1.1 Electronic states of diatomic molecules

The Born-Oppenheimer approximation allows the electronic structure of a molecule to be analysed independently of the nuclear motion. This is because the mass of the electrons is much smaller than that of the nuclei, resulting in the electronic motion being much faster than the nuclear motion. Therefore, the electronic states of YbF can be described independently of those related to nuclear motion.

Atomic quantum numbers, such as the principal quantum number, n , orbital angular momentum quantum number, l , and the projections of orbital and spin angular momentum, m_l and m_s , are used to describe the state of an atom. These quantum numbers describe the allowed energy levels and corresponding wave functions of an electron in an atom. However, these atomic quantum numbers are insufficient to describe the behaviour of electrons in a molecule. In a molecule, electrons interact with multiple nuclei, and each nucleus attracts the electrons with a different Coulombic potential. This leads to the electrons being distributed in the molecule in a manner that cannot be described by atomic quantum numbers alone.

When a diatomic molecule is formed, the two nuclei create a strong electric field between them. This electric field affects the motion of the valence electron, whose orbital angular momentum, $\mathbf{L}$, strongly couples to it. As a result, the electron's orbital angular momentum precesses rapidly around the internuclear axis, $\hat{n}$, making it impossible to use the orbital angular momentum as a good quantum number to describe the behaviour of the electron. However, the electron's orbital

angular momentum component along the internuclear axis, known as Λ , remains a good quantum number. This is because the internuclear axis defines a preferred direction in space. The Λ quantum number can take on values $-L, -L+1, \dots, L-1, L$. It is denoted using Greek characters, such as Σ, Π, Δ , for $|\Lambda| = 0, 1, 2, \dots$, in a manner similar to the spectroscopic notation for atoms.

In diatomic molecules, states with nonzero Λ correspond to electronic orbitals whose angular momentum vector has a component along the molecular axis. Due to the axial symmetry of the molecule, there are two possible orientations of the vector with respect to the axis, resulting in a two-fold degeneracy of the state. However, various interactions can lift this degeneracy by coupling the electronic state to the molecule's rotational or spin angular momentum. This is known as Λ -doubling and it leads to a splitting of the energy levels associated with the two possible orientations of the angular momentum vector, resulting in two distinct states that differ in energy. The magnitude of the Λ -doubling splitting depends on the coupling strength between the electronic and rotational angular momenta.

Just like in atoms, there is a spin-orbit interaction in molecules which couples the orbital angular momenta of the electrons to their spins. When the spin-orbit interaction is strong, the total electronic angular momentum along the internuclear axis, denoted by Ω , becomes a good quantum number. The projection of the spin angular momentum vector, S , onto the internuclear axis, denoted by Σ , also becomes a good quantum number. The possible values of Σ are $-S, -S+1, \dots, S-1, S$, with a multiplicity of $2S+1$, where S is the magnitude of the spin angular momentum. The sum of Σ and Λ then defines the total electronic angular momentum along the internuclear axis, $\Omega = \Lambda + \Sigma$. These quantities are combined to create a diatomic molecule's term symbol for a $\Lambda \neq 0$ state, $^{2S+1}\Lambda_{\Omega}$.

When the Λ quantum number equals zero, there is no spin-orbit coupling, so Σ and Ω are not well-defined. The electronic orbital angular momentum vector is perpendicular to the internuclear axis and, as a result, the molecule's orientation around the internuclear axis does not affect the electron's motion, and the state is not degenerate. However, to fully describe the $\Lambda = 0$ state, it is necessary to consider reflection through the internuclear axis. This leads to the emergence of two states, Σ^+ and Σ^- , which are distinguished based on the symmetry of the electronic wavefunc-

tion upon reflection through a plane containing the nuclei. The labels + and – correspond to the wavefunction being unchanged or changed in sign, respectively, upon reflection.

Each electronic state of a diatomic molecule is given a unique symbol to identify it. The electronic ground state is always labelled X. Electronic excited states are labelled alphabetically, A, B, C.... The electronic ground state of YbF is a $^2\Sigma^+$ state. The first electronically excited state is $^2\Pi$ which is split by the spin-orbit interaction into a $^2\Pi_{1/2}$ and a $^2\Pi_{3/2}$ with the $^2\Pi_{1/2}$ having the lower energy. In this work, the only electronic states used are $X^2\Sigma^+$ and $A^2\Pi_{1/2}$.

2.1.2 Rotational and vibrational states of diatomic molecules

YbF is a diatomic molecule that exhibits rotational and vibrational motion. In the following section, the rotational and vibrational states of YbF are explored using simple models and then further elaborated upon by examining higher-order contributions.

Modelling YbF as a rigid rotor

A diatomic molecule can be visualised as a rigid rotor, where the rotational motion is due to its spinning around the centre of mass, which, for a diatomic molecule, is on the internuclear axis. Rotation around the internuclear axis is negligible [50]. However, the molecule's rotation around a vector perpendicular to the internuclear axis, as shown in figures 2.1a and 2.1b, is not negligible. The rotational inertia is $I = \mu x_0^2$, where $\mu = m_1 m_2 / (m_1 + m_2)$ is the reduced mass of the two nuclei and x_0 is the equilibrium separation of the nuclei along axis x . Classically, the rotation energy, E_R , of the rigid rotor is $E_R = L^2 / 2I$, where L is the angular momentum of the system about the axis of rotation, and quantisation of L gives $L^2 = \hbar^2 R(R+1)$, where R is the (quantised) rotation quantum number $R = 0, 1, 2, \dots$. We then arrive at the equation for the rotation energy of a rigid rotor:

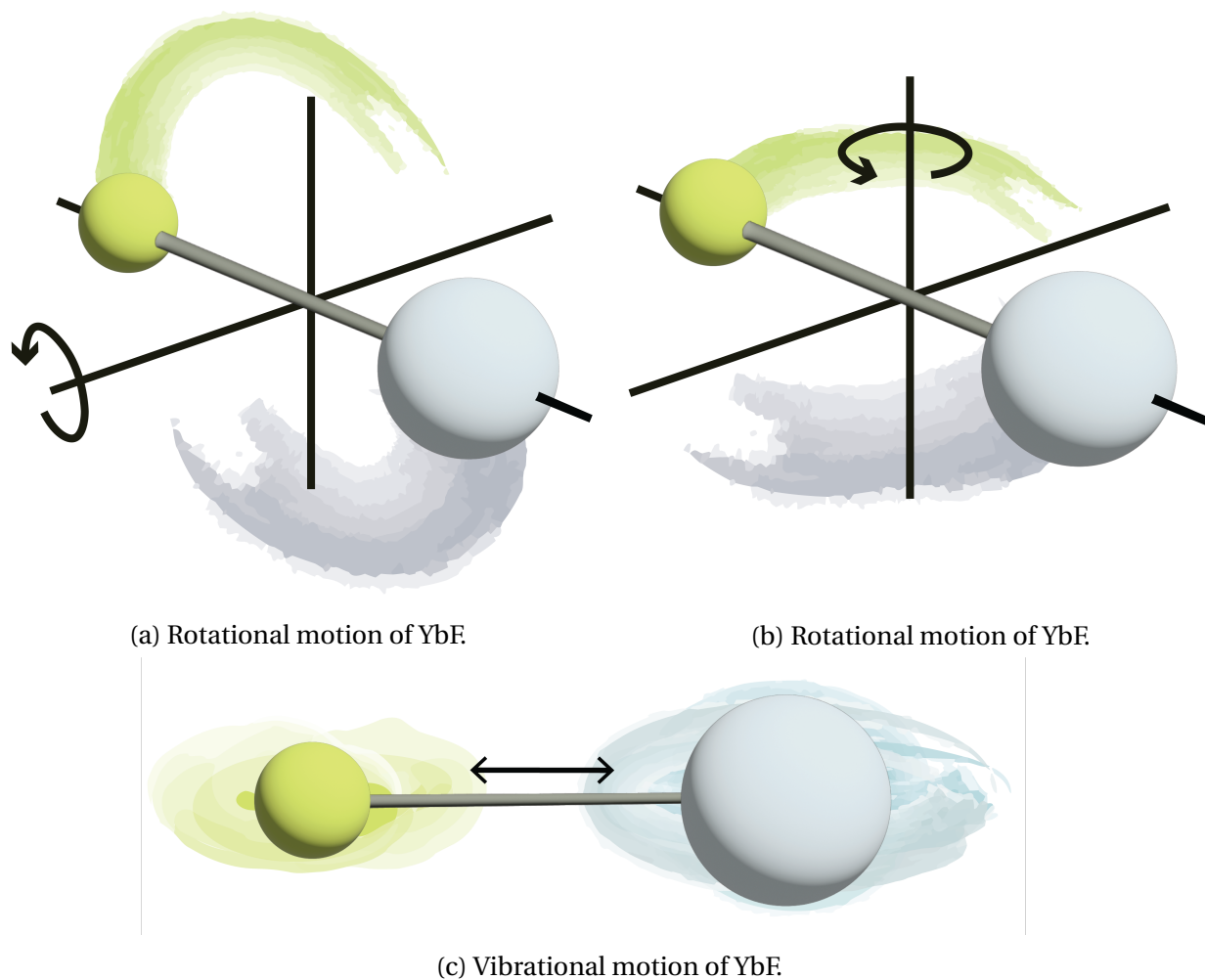


Figure 2.1: Illustration of nuclear motion in ytterbium monofluoride. Lime green spheres represent the fluorine nucleus, while silver spheres represent the ytterbium nucleus. It is important to note that these diagrams are not to scale, and the centre of mass is approximately 18 times closer to the ytterbium nucleus than the fluorine nucleus.

$$E_{\text{rot}} = \frac{h^2}{8\pi^2 I} R(R+1) \quad (2.1)$$

$$= BR(R+1) \quad (2.2)$$

where B is the rotational constant, $B = h^2/8\pi^2 I$.

Modelling YbF as a harmonic oscillator

The stretching and compression of the bond between the two atoms give rise to the vibrational motion of a diatomic molecule, as depicted in figure 2.1c. The potential energy of a diatomic molecule can be approximated as a simple harmonic oscillator, $V(r) = \frac{1}{2}k(x-x_0)^2$, where the bond length, x , is the displacement from the equilibrium position, x_0 , and the spring constant, k , is a measure of the bond strength. The Hamiltonian of the system is then,

$$\hat{H} = -\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial x^2} + \frac{1}{2}k(x-x_0)^2 \quad (2.3)$$

where μ is the reduced mass of the molecule. The allowed vibrational energies are then quantised and can be labelled using the vibrational quantum number ν , which takes integer values starting from zero. Each vibrational state corresponds to a specific energy level given by:

$$E_{\text{vib}} = \left(\nu + \frac{1}{2}\right) \hbar\omega_e \quad (2.4)$$

where $\omega_e = \sqrt{k/\mu}$ is the angular frequency of the harmonic oscillator, which depends on the electronic state of the molecule [50].

Higher-order corrections

The rotational and vibrational motions of a diatomic molecule are closely related and interact with each other. While the simple rigid rotor model can describe the rotational motion, higher-order corrections are needed to account for the centrifugal stretching of the bond as the molecule rotates. Similarly, the vibrational motion can be described by the simple harmonic oscillator model, but the potential energy curve deviates from a perfect parabolic shape, leading to higher-order corrections to the vibrational energy levels [50, 51]. Therefore, a more accurate potential is required to derive corrections to the simple vibration and rotation models that goes beyond the simple harmonic oscillator approximation. The Dunham potential is one such potential, obtained by adding higher-order terms to the potential energy expression [3, 52–54]. By using this potential, we can derive the following definitions for the rotational and vibrational energies:

$$E_{\text{rot}}(\nu, R) = B(\nu)R(R+1) - D(\nu)R^2(R+1)^2 + \dots, \quad (2.5)$$

$$E_{\text{vib}}(\nu) = \hbar\omega_e(\nu + 1/2) - \hbar\omega_e x_e(\nu + 1/2)^2 + \dots \quad (2.6)$$

Here, $D(\nu)$, the centrifugal distortion, depends on the molecule's vibrational state, $B(\nu)$ is a correction to the rotational constant, and x_e is a positive constant called the anharmonicity constant [53, 54].

Equation 2.2 shows that a rigid rotor's energy levels depend on the distance between the two nuclei, which appears through the inertia term in the rotational constant. However, when the atoms vibrate, this distance changes, and so does the rotational energy. Therefore, the rotational energy depends on the molecule's vibrational state. To account for this, the rotational constant becomes dependent on the vibrational quantum number:

$$B(\nu) \equiv B_e - \alpha_e \left(\nu + \frac{1}{2} \right) \quad (2.7)$$

The correction factor α_e is $\alpha_e = B_e^2/(\hbar\omega_e)$, where B_e is the equilibrium rotation constant [54].

In addition, the centrifugal distortion correction, $D(\nu)$, is another crucial correction affecting diatomic molecules' rotational energy levels. When a molecule rotates, the centrifugal force can cause the nuclei to move apart, elongating the bond length. This effect becomes more significant at higher rotation speeds and must be considered when analysing a diatomic molecule's rotational energy levels.

2.1.3 Hyperfine and spin-rotation interactions of YbF

Hyperfine interactions arise from the nuclear spin and the electron angular momenta coupling. The hyperfine interaction is typically weaker than other types of interactions but still larger than the natural linewidth of the transition used in this work. This section discusses the hyperfine interactions in the X and A states of YbF. We will use the following angular momenta here and throughout. $\vec{L}$ is the total electronic orbital angular momentum, $\vec{S}$ is the total electronic spin, $\vec{R}$ is the rotational angular momentum of the molecule, $\vec{I}$ is the spin of the fluorine nucleus, $\vec{N} = \vec{L} + \vec{R}$, $\vec{J} = \vec{N} + \vec{S}$ and $\vec{F} = \vec{J} + \vec{I}$.

Couplings in the ground electronic state in YbF

Parameter (units)	Value ($\nu = 0$)	Value ($\nu = 1$)
γ_0 (MHz)	-13.42400(16)	-33.8118(7)
γ_1 (kHz)	3.9823(11)	4.323(6)
γ_2 (mHz)	-25(1)	-28(9)
b_0 (MHz)	141.7956(5)	139.89(4)
b_1 (kHz)	-0.510	-0.7(4)
c (MHz)	85.4026(14)	86.75(5)
C (kHz)	20.38(13)	18.3(1)

Table 2.1: Table showing the spin-rotation and hyperfine parameters for the vibrational states $\nu = 0$ and $\nu = 1$ of the $X^2\Sigma^+$ electronic ground state in YbF. Parameters were obtained from Sauer et al. (1996) [55].

An effective Hamiltonian that describes some of the internal couplings of the ^{174}YbF isotopologue in its electronic ground state, $X^2\Sigma^+$, is presented in equation 2.8 [3, 55–57]. Notably, this Hamil-

tonian has no spin-orbit coupling since there is no orbital angular momentum and, therefore, $\vec{N} = \vec{J} - \vec{S} = \vec{R}$, so N and R are interchangeable.

$$H = \gamma \vec{N} \cdot \vec{S} + b \vec{I} \cdot \vec{S} + c I_z S_z + C \vec{I} \cdot \vec{N}. \quad (2.8)$$

The first term of the Hamiltonian, $\gamma \vec{N} \cdot \vec{S}$, describes the spin-rotation interaction. When a diatomic molecule rotates, it generates a magnetic field that can interact with the electron's magnetic moment associated with its spin. [58]. The spin-rotation constant, γ , strongly depends on the value of N , and can be expressed as an expansion in powers of $N(N+1)$:

$$\gamma = \gamma_0 + \gamma_1 N(N+1) + \gamma_2 (N(N+1))^2 + \dots, \quad (2.9)$$

where the coefficients γ_i decrease with increasing powers of $N(N+1)$. Table 2.1 lists the γ_i values for the $\nu = 0$ and $\nu = 1$ vibrational states of ^{174}YbF [55].

The other terms in equation 2.8, $b \vec{I} \cdot \vec{S}$, $c I_z S_z$ and $\vec{I} \cdot \vec{N}$, describe the hyperfine interaction between the nuclear spin and the rotational and electronic spin angular momenta. Here, it should be noted that subscript z refers to the direction of the internuclear axis. The hyperfine parameters, b , c and C , are defined in table 2.1, where b is parameterised in the same manner as γ in equation 2.9:

$$b = b_0 + b_1 N(N+1). \quad (2.10)$$

Our experiments consider the first three rotational states, with $N = 0, 1$, and 2 . Since the hyperfine interactions in these states are stronger than the spin-rotation coupling, we describe the states using a basis that couples the electronic spin and nuclear spin angular momenta, denoted as $\vec{G} = \vec{I} + \vec{S}$. Then, we couple the rotation and spin angular momenta to form $\vec{F} = \vec{G} + \vec{N}$. The ^{174}YbF

isotopologue has a nuclear angular momentum of $1/2$, so the total spin angular momentum can take values of $G = 0, 1$. The combination with the ground state angular momentum $N = 0$ then gives $F = 0, 1$. The combination with higher rotational states gives the total angular momenta $F = N - 1, N_l, N_h$, and $N + 1$, where l and h refer to the lower and higher energy states. Each of the F states has $(2F + 1)$ -fold degeneracy, which can be lifted by the Zeeman effect.

Couplings in the first excited electronic state of YbF

Here, we focus on the $J' = 1/2$ state of the first electronically excited state of YbF, denoted as $A^2\Pi_{1/2}$. As discussed above, Λ -doubling splits this level into a pair of states with opposite parity, conventionally denoted as $|e\rangle$ and $|f\rangle$ [49, 59]. These are:

$$|e\rangle = \frac{1}{\sqrt{2}} (|\Omega' = 1/2\rangle + |\Omega' = -1/2\rangle), \quad (2.11)$$

$$|f\rangle = \frac{1}{\sqrt{2}} (|\Omega' = 1/2\rangle - |\Omega' = -1/2\rangle). \quad (2.12)$$

The $\binom{e}{f}$ states have parity $(+ -)(-1)^{J-1/2}$ [3, 49, 59]. The hyperfine interaction further splits the states into two hyperfine states, $F' = 0, 1$. This hyperfine splitting is smaller than the natural linewidth of the A state, so we often neglect it [49, 56].

2.1.4 Perturbations in the first excited vibrational state of YbF's A state

The $A^2\Pi_{1/2}$ state of YbF is closely matched in energy to another YbF state, which has $\Omega = 1/2$ and vibrational and rotational constants that are consistent with the $4f^{13}6s^2$ configuration [59]. This state is designated as [18.6]0.5, where $[10^{-3}T_0]\Omega$ is a naming convention used for other lanthanide molecules and references the lowest energy state T_0 (in cm^{-1}) [59, 60]. The excited vibrational state of $A^2\Pi_{1/2}$ with $v' = 1$ is strongly perturbed by the [18.6]0.5 state, resulting in its splitting into admixture states, labelled as [557] and [561], which refer to the energy difference (in THz) from the

ground state $X^2\Sigma^+(\nu = 0)$. Further details on the radiative branching ratios of these levels can be found in [60], while measurements of the hyperfine structure are presented in [56].

2.1.5 Gross structure of YbF

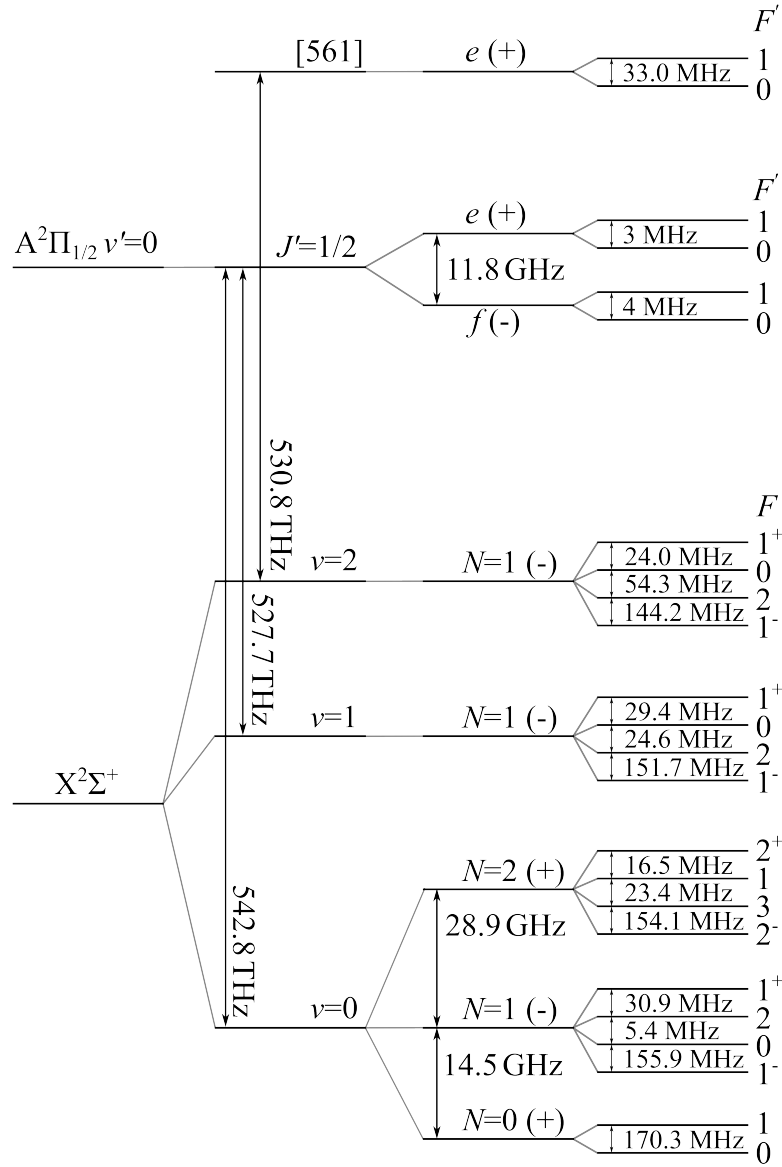


Figure 2.2: Energy level diagram illustrating the key energy levels of ^{174}YbF that are relevant for understanding this thesis, with spectroscopic information obtained from measurements published in Sauer et al. (1996) [55].

The energy level diagram presented in figure 2.2 illustrates the gross structure of ^{174}YbF relevant to this work. Starting with the electronic configurations, discussed in section 2.1.1, the diagram shows their splitting into various vibrational and rotational states, as explained in section 2.1.2,

followed by the hyperfine coupling-induced splitting into the F states. The electronically excited state further splits into the $|e\rangle$ and $|f\rangle$ states, as described in section 2.1.3. Additionally, due to its unique behaviour, the admixture state [561] is depicted separately from the $A^2\Pi_{1/2}$ state.

2.1.6 Transitions

In this experiment, we use several electric dipole (E1) transitions in YbF. The majority of the transitions occur between the $X^2\Sigma^+$ and $A^2\Pi_{1/2}(\nu' = 0)$ states. However, we also consider microwave transitions between rotational states and transitions from the $X^2\Sigma^+(\nu = 2)$ state to the [561] state. The following section provides an overview of all the transitions that are relevant to this study.

Selection rules and rotationally closed transitions

Let's start with the transitions between the $X^2\Sigma^+$ and $A^2\Pi_{1/2}$ states. Specific selection rules govern these transitions. The total angular momentum change can be $\Delta F = 0, \pm 1$, while the projection of F onto the internuclear axis may change by $\Delta m_F = 0, \pm 1$. If F does not change ($\Delta F = 0$), then its projection must change ($\Delta m_F = \pm 1$). Additionally, the total angular momentum without nuclear spin can change by $\Delta J = 0, \pm 1$, and the state's parity must also change [61]. However, there are no selection rules regarding the vibrational quantum number, ν .

When the molecule is excited to the positive parity component of the $A^2\Pi_{1/2}(J' = 1/2)$ state, the selection rules for parity and angular momentum restrict the possible decay channels to just a single rotational state, $N = 1$, of the electronic ground state forming a rotationally closed set of transitions that are used for laser cooling YbF molecules. Similarly, the decay from the odd parity f state is only allowed if the final state has even parity, i.e., $N = 0, 2, \dots$. Once again, the selection rules limit the decay, forming a rotationally closed set of transitions between the $N = 0$ and $N = 2$ ground states.

Overview of transition branches and relevant transitions in the experiment

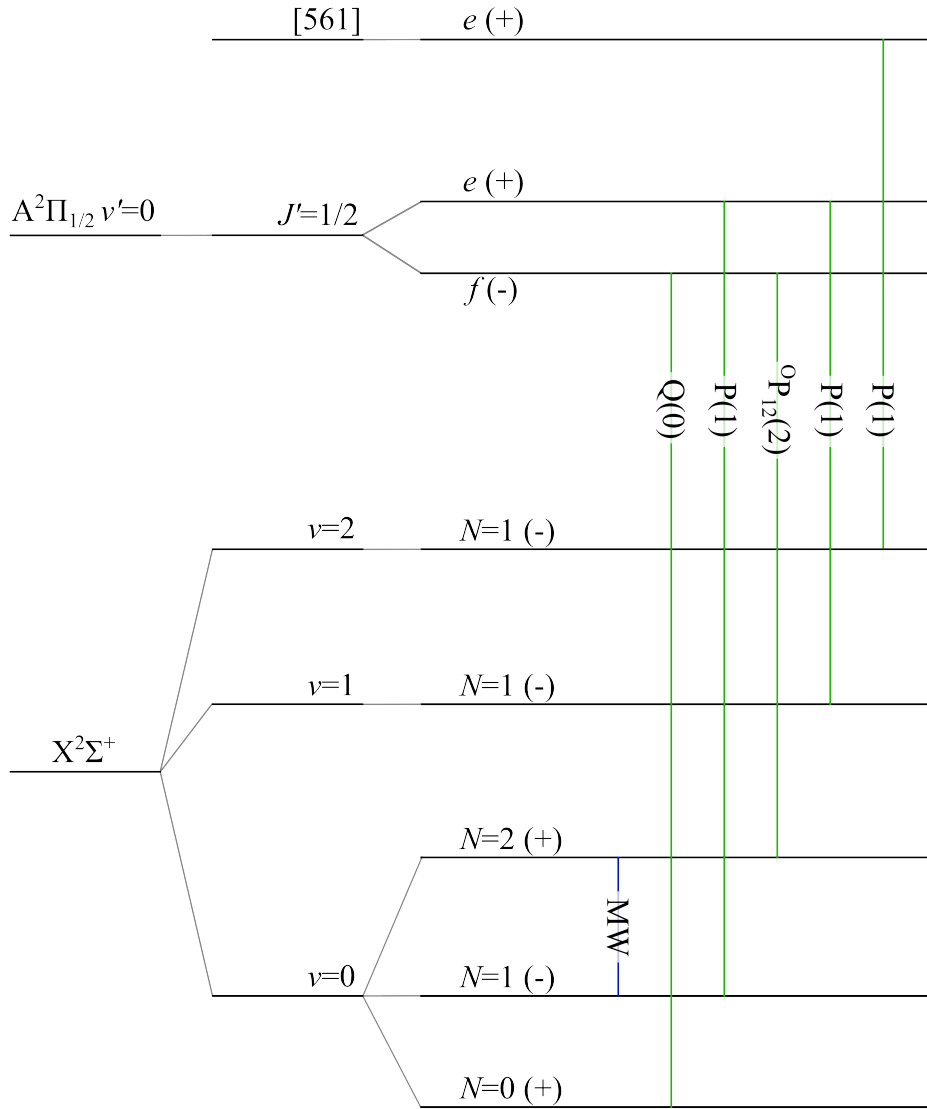


Figure 2.3: Energy level diagram showing the relevant groups of transitions of YbF for understanding this thesis. The P and Q branches between electronic states are indicated as $P(N)$ and $Q(N)$, respectively, where N signifies the rotational state of the ground state of the transition. Microwave (MW) transitions between rotational states are also included. The transition indicated from $N = 2$ falls outside the categorization into P , Q and R lines and is labelled $^0P_{12}(2)$.

When deriving the energy of transitions using the equations outlined in section 2.1.2, the transitions can be classified into three groups [62, 63]. The first group is called the R-branch, which comprises transitions where $\Delta J = +1$, meaning that the total angular momentum (excluding nuclear spin) in the excited state is one unit more than that in the ground state, i.e., $J' = J + 1$. The R-branch transitions typically have the highest energy of the three groups. The second group is

the Q-branch, which involves transitions with $\Delta J = 0$. Because the rotational constants are similar in the ground and excited states, the Q transitions have similar energies for all values of J . Therefore the lines cluster together, and the spectrum is congested in this region. The third group is the P-branch, which are the transitions with $\Delta J = -1$. The P-branch transitions have the lowest energy among the three groups.

The $P(1)$ optical transitions are used for laser cooling, as discussed in chapter 4. A combination of microwave and optical transitions are used to optically pump the population between rotational states, as discussed in chapter 6.

Vibrational transitions and branching ratios

When considering vibrational states, it is necessary to take into account the Franck-Condon factors (FCFs). The FCF determines the relative probability for the transition between the vibrational state ν of one electronic state and the vibrational state ν' of a different electronic state [50]. This probability can be calculated as the square of the overlap between these two vibrational wavefunctions, expressed as $|\langle \nu' | \nu \rangle|^2$.

For optimal laser cooling, the ideal scenario is to have a laser cooling transition with an FCF of one. However, this is not the case with the cooling transition used in our experiment, which has an FCF of $|\langle \nu' = 0 | \nu = 0 \rangle|^2 = 0.928$. This means that on average, $\Gamma = 1/(1 - |\langle \nu' = 0 | \nu = 0 \rangle|^2) \approx 14$ photons are scattered before a molecule decays to a higher vibrational state. Repumping of the higher vibrational states is crucial for our laser cooling scheme, as discussed in chapter 4. Table 2.2 provides the relevant FCFs for transitions between the $X^2\Sigma^+$ and $A^2\Pi_{1/2}(\nu' = 0)$ and [561] states.

	$\nu = 0$	$\nu = 1$	$\nu = 2$	$\nu = 3$
$\nu' = 0$	0.93(3)	0.066(3)	0.0030(5)	$0 \pm 5 \times 10^{-4}$
[561]	0.028(3)	0.89(1)	0.078(2)	$0 \pm 2 \times 10^{-3}$

Table 2.2: The Franck-Condon factors (FCFs) for transitions between the $X^2\Sigma^+$ vibrational states $\nu = 0-3$ and the $A^2\Pi_{1/2}$ and [561] states. The FCFs for $A^2\Pi_{1/2}$ - $X^2\Sigma^+$ transitions were obtained from Smallman et al. (2014) [60], while those for [561]- $X^2\Sigma^+$ transitions were obtained from Zhuang et al. (2011) [64].

2.2 Interactions of ytterbium monofluoride with external fields

The YbF molecules' interactions with external electric and magnetic fields are essential to our experiment, serving a critical role in laser cooling, which is discussed in chapter 4. These interactions are also central to the YbF eEDM measurement technique, as described in chapter 1, and are integral to the operation of the magnetic lens discussed in chapter 5.

2.2.1 Interactions with magnetic fields

When a magnetic field $\vec{B}$ is applied to a YbF molecule, the valence electron will interact with it via the Zeeman effect. In the weak field limit, the Zeeman splitting is small compared to the hyperfine splitting. In this case, we can describe the state of the molecule using the Hamiltonian, $\hat{H} = \hat{H}^{(0)} + \hat{H}_{\text{FS}} + \hat{H}_{\text{ZE}}$, where $\hat{H}^{(0)}$ is the bare Hamiltonian, $\hat{H}_{\text{FS}}$ is the fine structure Hamiltonian, and $\hat{H}_{\text{ZE}}$ is the Zeeman effect Hamiltonian. Since the energy change associated with the Zeeman effect is less than that of the fine structure, we treat $\hat{H}_{\text{ZE}}$ as a perturbation to the $(\hat{H}^{(0)} + \hat{H}_{\text{FS}})$ Hamiltonian.

In the ground state $X^2\Sigma^+$ of YbF, there is no orbital angular momentum to consider. Therefore, $\hat{H}_{\text{ZE}}$ depends only on the dot product of the electron spin angular momentum, $\vec{S}$, and magnetic field:

$$\hat{H}_{\text{ZE}} = g_s \mu_B \vec{S} \cdot \vec{B} \quad (2.13)$$

$$= g_s \mu_B S_z B_z. \quad (2.14)$$

In equation 2.14, S_z and B_z are projections onto the internuclear axis z , so the interaction depends on the alignment of the molecule relative to the applied field. Here, g_s is the electron spin g-factor, which is approximately 2.002, and μ_B is the Bohr magneton. Other contributions to the Zeeman effect, coming from the nuclear magnetic moment and the rotation of the molecule, are three orders of magnitude smaller and are neglected here.

The first-order shift in the energy due to the Zeeman effect of the $X^2\Sigma^+$ state is then given by per-

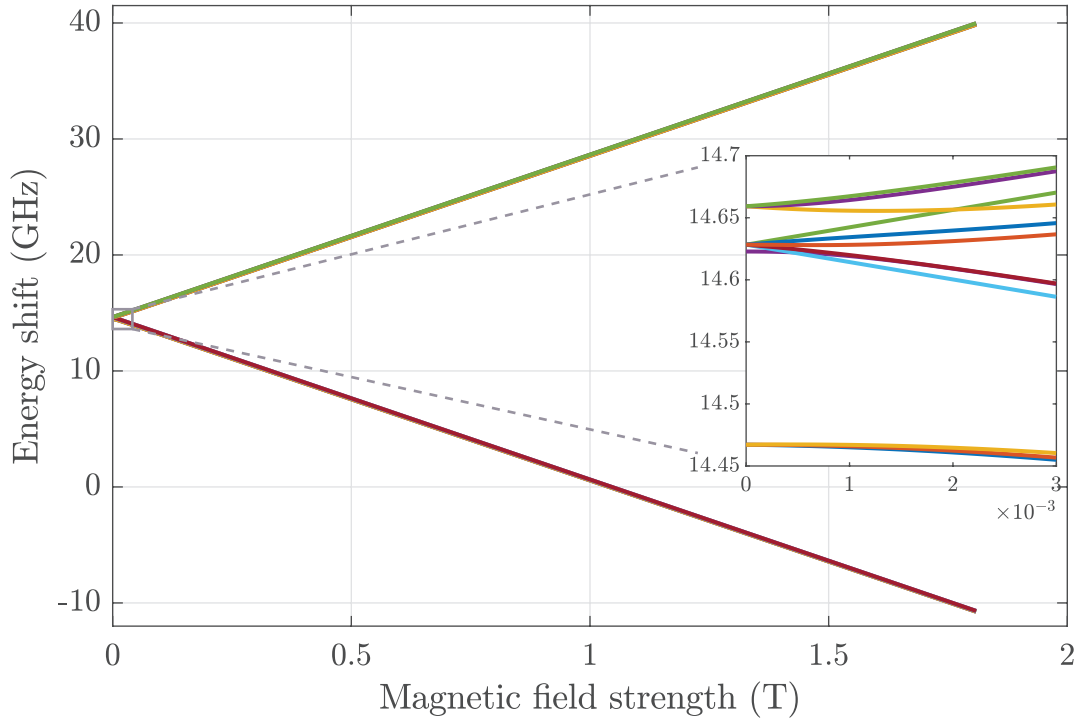


Figure 2.4: Simulated energy shifts of the m_F states in the $X^2\Sigma^+(N=1)$ state of YbF due to the Zeeman effect, as a function of applied magnetic field strength. The inset plot shows the weak field dependencies of the energy shifts, which are more complex due to the different m_F and g_F combinations.

turbation theory as the expectation value of H_{ZE} , with the result:

$$\Delta E_{ZE} = g_F \mu_B m_F B_z. \quad (2.15)$$

The Landé g-factors, g_F , for each of the F states were calculated by J. Devlin and can be found in appendix B.3 of his thesis [2].

The strong field regime is reached for the ground state when the Zeeman shift is much larger than the hyperfine splittings. In this case, the individual angular momenta, $\vec{N}$, $\vec{S}$ and $\vec{I}$ decouple, and their individual projections onto the z-axis are all good quantum numbers. In this case, the expectation value of H_{ZE} is simply $g_s \mu_B B m_S \approx \pm \mu_B B$, since $g_s \approx 2$ and $m_S = \pm 1/2$.

The importance of considering both weak and strong applied fields is two-fold. In eEDM inter-

ferometric measurements, a weak magnetic field is applied to the molecules, and the precession of the superposition state is used to measure the eEDM. Therefore, it is essential to understand the weak field dependence of the Zeeman shift. On the other hand, the high field limit must be considered for molecules propagating through a magnetic lens, as discussed in chapter 5, where the magnetic field strength can be as high as 1.4 T.

Figure 2.4 displays the energy shifts of the m_F states in $X^2\Sigma^+(N=1)$ upon exposure to an applied magnetic field. The zero-field energy shift is primarily due to the ~ 14.5 GHz rotational splitting, with the $(N=0, F=0)$ state at the origin. At high field strengths, six of the m_F states exhibit an energy shift of approximately $-\mu_B B$, while the other six states have a shift of $+\mu_B B$. These two sets of lines in the figure overlap. At a field strength of ~ 1.4 T, which is the magnetic field strength experienced by $N=1$ molecules propagating through the magnetic lens in chapter 5, the Zeeman shifts are $\pm\mu_B B = \sim 20$ GHz. The inset plot shows the weak field dependence, which follows a more complex trend due to the various m_F and g_F combinations.

2.2.2 Interactions with electric fields

The AC Stark shift is a phenomenon that occurs when a molecule is exposed to a non-stationary external electric field, such as the electric field of a laser beam. This field induces transitions between different states of the molecule, as well as causing energy level shifts. In the simplest case of a two-level system, the ground state g and the excited state e are addressed by an electromagnetic wave with a frequency ω near the transition resonance frequency ω_0 . The Hamiltonian for this system has off-diagonal terms, which contain the Rabi frequency Ω . As a result, the energies of g and e are shifted equally and oppositely:

$$\Delta E = \pm \frac{\hbar\Omega^2}{4\delta}, \quad (2.16)$$

where $\delta = \omega - \omega_0$ is the detuning of the light from the transition resonance. This effect is some-

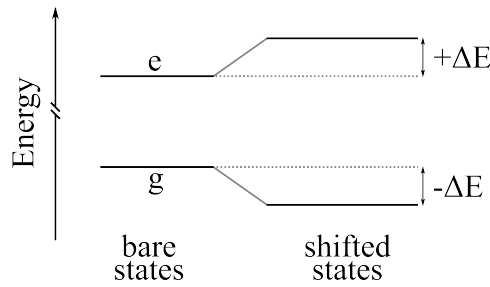


Figure 2.5: Energy level diagram illustrating the energy shifts of a two-level system consisting of a ground state g and an excited state e under the influence of a laser field. The laser field causes the energy levels of the two states to shift in opposite directions by $\pm\Delta E$.

times referred to as the "light shift" due to its dependence on the intensity of the light, which is proportional to Ω^2 [65]. Figure 2.5 illustrates this light shift, showing how the two energy levels are shifted away from one another, increasing the transition frequency when the light is red-detuned.

The AC Stark shift is an important effect in laser cooling and trapping. Laser cooling is achieved by tuning the laser frequency to the blue side of the atomic resonance so that the AC Stark shift pushes the excited state energy level downwards, as is discussed in section 2.3. Conversely, the trapping of atoms in an optical trap requires the laser frequency to be red-detuned from the atomic resonance to produce a restoring force.

2.3 Fundamentals of laser cooling for atoms and molecules

In this section, I explore the fundamental concepts involved in the laser cooling of atoms and molecules. It begins with a concise overview of the historical development of laser cooling in section 2.3.1. Following that, in section 2.3.2, I explain some of the fundamental light-matter interactions that form the basis of laser cooling mechanisms. Finally, in sections 2.3.3 and 2.3.4, we delve into the heart of the matter, as I describe the mechanisms of Doppler and sub-Doppler laser cooling.

2.3.1 A historical overview of laser cooling

The idea of radiation pressure on matter can be traced back at least to the early 17th century when Kepler proposed that comet tails point away from the Sun due to an interaction with the Sun's light [66]. However, it was not until the turn of the 20th century that Lebedev demonstrated the force exerted on macroscopic objects by light [67, 68]. The early 20th century saw significant activity in this field, particularly with the advent of Einstein's quantum theory of light [69]. The concept of radiation pressure was fundamental to the development of laser cooling, which relies on using laser light to slow down the motion of individual atoms and molecules.

Laser cooling is an innovative technique that enables scientists to slow down the motion of individual atoms to near-absolute-zero temperatures. The concept of laser cooling was first proposed by Schawlow and Hänsch in 1975 [70], and later independently by Dehmelt and Wineland in the same year [68, 71]. However, it wasn't until 1978 that the first successful experiments were conducted by Drullinger, Wineland, and Walls, who laser-cooled a cloud of magnesium ions confined in a Penning trap [72]. Doppler and sub-Doppler cooling techniques have made significant advancements since their initial development. In the mid-1980s, researchers designed various methods for confining atoms, such as magnetic traps, optical dipole traps, and magneto-optical traps (MOTs) [68, 73–75]. These new developments in atom confinement allowed for improved cooling mechanisms.

In the late 1980s, researchers discovered sub-Doppler cooling mechanisms [76, 77], which made it possible to cool atoms to even lower temperatures than previously possible. This discovery opened up new avenues of research in fields such as quantum computing [78] and atomic clocks [79], and has led to significant advancements in precision measurements of fundamental constants [1, 42, 48, 80], inertial sensing [81–83], and more. Today, Doppler and sub-Doppler cooling techniques are widely used in state-of-the-art scientific and technological applications, highlighting the importance of continued research and innovation in this field.

Laser cooling of molecules is a challenging task due to their complex internal energy structure, which includes rotational and vibrational states and associated internal interactions. However,

in the 21st century, the development of ultracold molecules was achieved. Some groups used the same approach as the start of atomic laser cooling, i.e., decelerating molecular beams via radiation pressure [84]. Others combined laser-cooled atoms to form ultracold bialkali molecules [85, 86]. The early 2010s witnessed significant progress in this field, with 2D and 3D MOTs created for various diatomic molecular species such as YO, SrF, and CaF [87–90]. Recently, the laser cooling of the first polyatomic molecule, YbOH, was demonstrated, with the temperature reaching below $600\text{ }\mu\text{K}$ [7]. Additionally, the successful trapping and cooling of CaOH in a MOT to $110(4)\text{ }\mu\text{K}$ [91] marks an important step forward in laser cooling of polyatomic molecules, suggesting the potential for cooling of other complex species in the future.

2.3.2 Light-matter interactions

In this section, I lay the groundwork for the laser cooling concepts covered in sections 2.3.3 and 2.3.4. I first introduce the concept of Rabi oscillations and their significance, followed by a discussion of the optical Bloch equations and their solutions, which underlie the physical phenomena that enable laser cooling. Lastly, I explore the scattering force that arises from photon absorption and emission.

Rabi oscillations

Rabi oscillations, also known as Rabi cycles or Rabi flops, describe the behaviour of a two-level quantum system in the presence of an oscillating electromagnetic field, such as a laser [61, 65]. For a plane wave, the electric field is given by $\vec{E}(\vec{r}, t) = E_0 \hat{e} \cos(kz - \omega_L t)$, where $\hat{e}$ is the unit polarisation vector, E_0 is the amplitude of the light field, and ω_L is the angular frequency of the electromagnetic wave.

In the case of a two-level atom with a ground state $|e\rangle$ and an excited state $|g\rangle$, the wavefunction is given by:

$$\Psi(\vec{r}, t) = c_e(t)\phi_e(\vec{r})e^{-i\omega_e t} + c_g(t)\phi_g(\vec{r})e^{-i\omega_g t}, \quad (2.17)$$

where ω_e and ω_g are the respective frequencies of the ground and excited states, and $\phi_e(\vec{r})$ and $\phi_g(\vec{r})$ are the spatial wave functions.

The Rabi frequency, denoted by Ω , characterises the coupling strength between the two-level system and the electromagnetic field. It is defined as:

$$\Omega \equiv -\frac{E_0}{\hbar} \langle e|d|g \rangle, \quad (2.18)$$

where d is the electric dipole moment operator and $\langle e|d|g \rangle$ is the matrix element of the dipole operator between the ground and excited states.

After solving the time-dependent Schrödinger equation, we obtain two coupled differential equations for the amplitudes $c_e(t)$ and $c_g(t)$, with Rabi frequency Ω as a parameter:

$$i \dot{c}_e = c_g \Omega \cos(\omega_L t) e^{i\omega_{eg} t}, \quad (2.19)$$

$$i \dot{c}_g = c_e \Omega^* \cos(\omega_L t) e^{-i\omega_{eg} t}, \quad (2.20)$$

where $\omega_{eg} = \omega_e - \omega_g$ is the frequency difference between the two levels.

The coupled differential equations 2.19 and 2.20 describe the time-dependent behaviour of the probability amplitudes of the two states in a two-level system. Assuming initial conditions $c_g(t) = 1$ and $c_e(t) = 0$, the solutions are given by equations 2.21 and 2.22. These solutions show that the system oscillates between the excited and ground states at a frequency of $\Omega' = \sqrt{\Omega^2 + \delta^2}$, where $\delta = \omega_L - \omega_{eg}$ is the frequency detuning of the plane wave from the transition frequency.

$$c_e(t) = -i \frac{\Omega}{\Omega'} e^{-i\frac{\delta t}{2}} \sin \frac{\Omega' t}{2}, \quad (2.21)$$

$$c_g(t) = e^{i\frac{\delta t}{2}} \left(\cos \frac{\Omega' t}{2} - i \frac{\delta}{\Omega'} \sin \frac{\Omega' t}{2} \right). \quad (2.22)$$

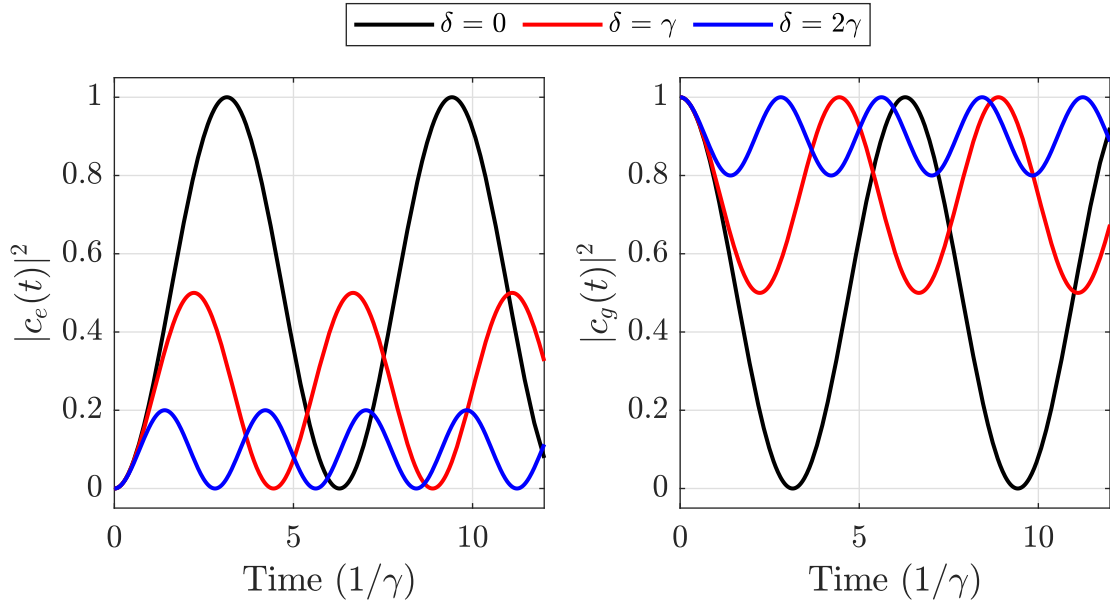


Figure 2.6: Time evolution of the probability densities for the ground and excited states of a two-level system in the presence of an electromagnetic wave. The figure shows the squares of the probability amplitudes, $|c_e(t)|^2$ and $|c_g(t)|^2$, as functions of time for three different detunings.

Figure 2.6 shows the squares of the probability amplitudes, $|c_e(t)|^2$ and $|c_g(t)|^2$, as functions of time for three different detunings. As the detuning increases, the oscillations' amplitude decreases while the oscillations' frequency increases. It is important to note that, in this model, spontaneous emission is not accounted for, so the atoms continue to oscillate between states indefinitely.

Optical Bloch equations

To account for spontaneous emission, one must consider the vacuum fluctuations of the light field, which requires quantisation of the field. Instead of directly quantising the field, we can include factors related to spontaneous emission, following the same method as in [65]. However, it should be noted that quantisation of the electromagnetic field would yield the same result.

To include spontaneous emission, we need to use the density matrix instead of state amplitudes to determine the statistical state of the system. The density matrix, ρ , satisfies the Liouville–von Neumann equation, which governs how the density operator evolves in time. For a pure state, the density matrix can be expressed as a matrix with the state amplitudes. Applying the Liouville–von Neumann equation to the density matrix elements leads to four differential equations relating to the probability amplitudes.

Spontaneous emission can be accounted for by letting the off-diagonal elements decay exponentially with constant $\gamma/2$ and subtracting a factor of $\gamma\rho_{ee}$ from $\dot{\rho}_{ee}$ to describe spontaneous decay from the excited to the ground state. Since $\rho_{gg} + \rho_{ee} = 1$ is conserved, we must also add $\gamma\rho_{ee}$ to $\dot{\rho}_{gg}$. Thus, the following equations are obtained:

$$\frac{d\rho_{gg}}{dt} = \gamma\rho_{ee} + \frac{i}{2}(\Omega^*\tilde{\rho}_{eg} - \Omega\tilde{\rho}_{ge}) \quad (2.23)$$

$$\frac{d\tilde{\rho}_{ge}}{dt} = -\left(\frac{\gamma}{2} + i\delta\right)\tilde{\rho}_{ge} + i\frac{\Omega^*}{2}(\rho_{ee} - \rho_{gg}) \quad (2.24)$$

$$\frac{d\rho_{eg}}{dt} = -\left(\frac{\gamma}{2} - i\delta\right)\tilde{\rho}_{eg} + i\frac{\Omega}{2}(\rho_{gg} - \rho_{ee}) \quad (2.25)$$

$$\frac{d\rho_{ee}}{dt} = -\gamma\rho_{ee} + \frac{i}{2}(\Omega\tilde{\rho}_{ge} - \Omega^*\tilde{\rho}_{eg}) \quad (2.26)$$

The notation $\tilde{\rho}_{ge} = \rho_{ge}e^{-i\delta t}$ is used to take into account the time dependence of the state amplitudes. The first equation describes the population of the ground state, while the next two equations describe the coherences between the ground and excited states. The last equation describes the population of the excited state.

Steady state solutions, power broadening and saturation

The steady-state solution of the optical Bloch equations for the excited state population is

$$\rho_{ee} = \frac{s}{2(1+s)} = \frac{s_0/2}{1+s_0+(2\delta/\gamma)^2}, \quad (2.27)$$

where s is the saturation parameter and is defined as:

$$s \equiv \frac{s_0}{1+(2\delta/\gamma)^2}. \quad (2.28)$$

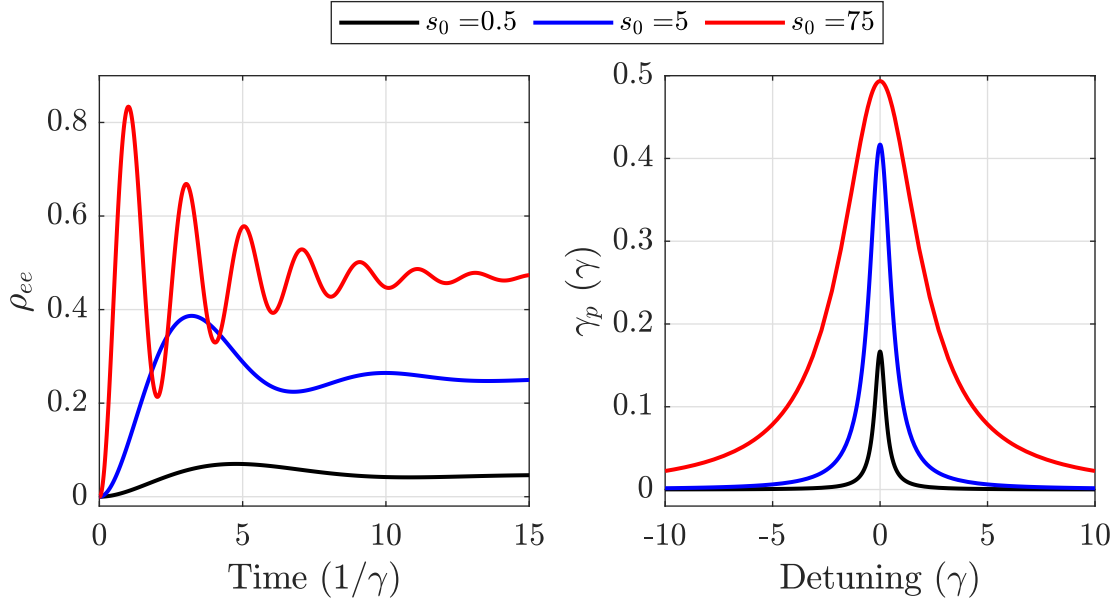


Figure 2.7: The left plot displays the time evolution of ρ_{ee} , calculated through numerical integration methods, for an initial population in the ground state, with $\delta = \gamma$. The right plot shows the scattering rate as a function of detuning. Both plots share the same legend, which shows three curves for each plot, corresponding to three on-resonance saturation parameters: $s_0 = 0.5$, 5, and 75.

Here, s_0 is the on-resonance saturation parameter, which is defined as $s_0 \equiv 2|\Omega|^2/\gamma^2 = I/I_s$. The intensity of the applied light is denoted by I , and the saturation intensity for the transitions is given by $I_s = \pi\hbar c/3\lambda^3\tau$, where λ is the wavelength of the transition and $\tau = 1/\gamma$ is the excited state lifetime.

In the low-intensity limit, the saturation parameter s is very small, resulting in most of the population remaining in the ground state. However, as the intensity of the applied light increases, the population splits almost evenly between the ground and excited states. This is because the excitation and decay rates are equal in the steady-state solution [65, 92]. Similarly, when the detuning is very large, s is small, and the probability of the system being excited is very small.

The photon scattering rate, γ_p , is the product of the decay rate and the probability of the system being in the excited state [61, 65, 92, 93]:

$$\gamma_p = \gamma\rho_{ee} = \frac{s_0\gamma/2}{1 + s_0 + (2\delta/\gamma)^2}. \quad (2.29)$$

The scattering rate can be rewritten as,

$$\gamma_p = \frac{s_0}{1 + s_0} \frac{\gamma/2}{1 + (2\delta/\gamma')^2}, \quad (2.30)$$

where $\gamma' = \gamma\sqrt{1 + s_0}$ is the power-broadened linewidth of the transition.

Figure 2.7 presents two plots that illustrate these concepts. The left plot shows the time evolution of ρ_{ee} as a function of the saturation parameter s_0 , calculated using numerical integration methods applied to the optical Bloch equations 2.23-2.26. The simulation assumes the system starts in the ground state and $\delta = \gamma$. As the intensity increases, and in turn, s_0 , the steady-state value of ρ_{ee} increases too, but not exceeding $\rho_{ee} = 0.5$. As the intensity increases, the linewidth of the transition broadens, as demonstrated in the right plot of Figure 2.7, where the scattering rate broadens with increasing saturation of the transition.

This power broadening of the transition has important implications for our experiments. When we tune our lasers, we need to reduce the laser power so that the transition we address does not broaden and obfuscate the transition's resonance. This consideration applies not only to the tuning of the lasers but also to any frequency-dependent measurements, such as those presented in chapter 6. The power broadening of the laser cooling transition is an important topic in chapter 4.

The scattering force (radiation pressure)

This section's final topic is the scattering force, also known as radiation pressure, that photons exert on a two-level system. When the system absorbs a photon, its momentum is entirely transferred to the system. The photon's momentum, $\mathbf{p}$, is related to the wave vector, $\mathbf{k}$, by the equation $\mathbf{p} = \hbar\mathbf{k}$. Upon emission of a photon, the two-level system loses momentum $\mathbf{p} = \hbar|\mathbf{k}|$ in the direction of the emitted photon. If the emission is stimulated, the emitted photon will propagate with the same wave vector as the absorbed photon, and the net momentum change for the system is zero. Unlike stimulated emission, the photons emitted through spontaneous emission can be emitted

in any random direction. As a result, the average momentum emitted by the two-level system is zero. However, in the direction of the wave's propagation, denoted by $\hat{k}$, the system accumulates momentum gains from the photons it absorbs, resulting in a net acceleration of the system in this direction [61, 65, 92, 93].

The force acting on the two-level system can be expressed as the rate of change of its momentum. The photon scattering rate, γ_p , is equation 2.30, and the system absorbs an average momentum of $p = \hbar k$ for each scattered photon. Thus, the scattering force, F , is given by:

$$F = p\gamma_p \quad (2.31)$$

$$= \hbar k \frac{s_0 \gamma / 2}{1 + s_0 + (2\delta / \gamma)^2}. \quad (2.32)$$

The scattering force is directly proportional to the scattering rate and behaves in the same manner as γ_p when the intensity of applied light is varied (see figure 2.7). When the intensity of applied light increases, the breadth of the force vs detuning peak widens, with the full width at half maximum being $\gamma' = \gamma \sqrt{1 + s_0}$ [93]. In the limit $I \rightarrow \infty$, the force reaches a maximum value of $F = \hbar k \gamma / 2$ [61, 93]. The spontaneous decay rate of the transition limits this maximum force.

2.3.3 Doppler cooling

In this section, I provide an overview of the fundamental principles of Doppler laser cooling within the framework of one-dimensional optical molasses. Although our experiment does not employ this specific cooling mechanism, it remains relevant because our blue-detuned laser cooling mechanism for YbF induces Doppler heating (as discussed in section 2.3.4). Towards the end of this section, I also examine the temperature limitations imposed by recoil from the scattering of photons and the Doppler cooling limit.

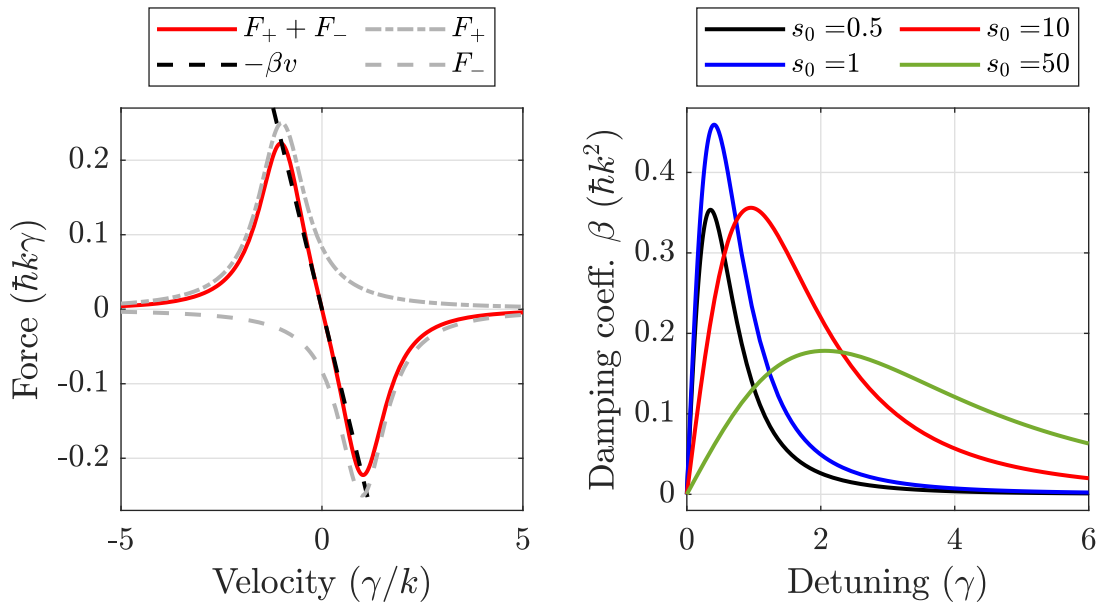


Figure 2.8: The left-hand plot illustrates the dependence of the scattering force on the velocity of a two-level system subjected to two counter-propagating waves, with $\Omega = \gamma/\sqrt{2}$ and $\delta = -\gamma$. The plot also displays the damping coefficient, β , of the combined forces at low velocity. The right-hand plot shows the damping coefficient as a function of detuning for different saturation intensities.

One-dimensional optical molasses

In section 2.3.2, we discussed the scattering force exerted on a two-level system by a single travelling wave. When an additional travelling wave, with the same detuning and intensity but propagating in the opposite direction, is included, the resulting forces from the two waves cancel out when $v = 0$, but do not cancel out when the atom is moving because of the Doppler shift. The total force F_T is given by $F_T = F_+ + F_-$, where F_+ and F_- are the forces exerted by the counter-propagating waves, as described in equations 2.33 [61].

$$F_{\pm} = \pm \hbar k \frac{s_0 \gamma / 2}{1 + s_0 + 4(\delta \mp \nu k)^2 / \gamma^2}, \quad (2.33)$$

Here, we have added the Doppler shift $\pm \nu k$ to the applied detuning. We can approximate the total force near $v = 0$ by a Taylor expansion of F_T . Combining expansion terms up to and including second order results in $F_T \approx -\beta v$, where β is the damping coefficient. It is given by [61, 65, 93]:

$$\beta = \hbar k^2 \frac{8s_0(\delta/\gamma)}{(1 + s_0 + 4(\delta/\gamma)^2)^2} \quad (2.34)$$

Figure 2.8 illustrates the velocity-dependent forces and damping coefficients of a two-level system subjected to counter-propagating waves with equal detunings $\delta = -\gamma$, i.e., red-detuned from the resonance frequency. The left-hand plot displays the separate forces as dashed lines and their sum as a solid red line. The force is positive when the velocity v is negative, causing deceleration, and negative when v is positive, still resulting in deceleration. At low velocities, the restoring force is strongest and can be well approximated by $F_T \approx -\beta v$, where β is the damping coefficient defined by the applied light intensity and detuning [65]. At the critical point of $v = 0$, the force vanishes. The right-hand plot in figure 2.8 shows the damping coefficient for different intensities. The plot illustrates how the damping coefficient increases until it reaches a maximum at an intermediate range of detunings and intensities. It is worth noting that if the applied light were blue-detuned instead, the descriptions of the force and velocity would be inverted, such that there would be velocity-dependent acceleration instead of deceleration.

The force acting on the atoms is always a damping force. When the interaction time between atoms and light is sufficiently long, their mean velocity will be damped to zero, regardless of their initial velocity. However, in practical scenarios, laser light has a finite width. If an atom moves out of the region illuminated by the light before its velocity reaches zero, it will no longer experience the damping force. The maximum velocity at which an atom's motion can be reduced to zero is called the capture velocity.

Here, we have considered only a single axis. However, the concept can be extended to three dimensions by introducing counter-propagating light in the other two orthogonal axes. This results in three-dimensional optical molasses, where the system experiences a velocity-dependent force that depends on its velocity in all three dimensions [61, 65, 93]. For the right detuning, this is a damping force.

Doppler cooling temperature limits

The momentum kicks that result from photon scattering lead to a random walk in momentum space [61]. This random walk can become significant when the velocity is low. This is where the temperature limit, known as the recoil temperature, T_r , comes into play [65]. T_r is defined by the equation:

$$k_B T_r \equiv \frac{\hbar^2 k^2}{2M}, \quad (2.35)$$

where k_B is Boltzmann's constant, and M is the mass of the particle being cooled. This equation is derived from the recoil velocity, $v_r = \hbar k / M$, which is the velocity imparted when a single photon is scattered [42, 61, 65, 93]. T_r is known as the recoil temperature limit and is typically a few microkelvins [65].

The recoil temperature limit given by Equation 2.35 cannot be achieved by using Doppler cooling alone because, at such low velocities, the heating rate due to spontaneous emission is usually greater than the cooling rate. The heating rate is $\dot{E}_{\text{heat}} = 4E_r \gamma_p$, where $E_r = M v_r^2 / 2$ is the recoil energy caused by a single photon, and γ_p is the scattering rate as defined in Equation 2.29 [61].

The cooling rate for low velocities can be calculated as the product of the cooling force and velocity, $\dot{E}_{\text{cool}} = F v = -\beta v^2$. By setting the heating and cooling rates equal in the steady-state solution, we obtain the root-mean-squared velocity as:

$$v_{\text{RMS}} = 2 \sqrt{\frac{\hbar^2 k^2 \gamma_p}{2M\beta}}. \quad (2.36)$$

When we equate this with the v_{RMS} for a one-dimensional thermal distribution, $\sqrt{k_B T / M}$, and solve for T in the limit $v \rightarrow 0$, we get:

$$T = -\frac{\hbar\gamma^2}{8k_B\delta} (1 + s_0 + (2\delta/\gamma)^2). \quad (2.37)$$

This equation is only valid for red-detuned light since we have assumed that there is a restoring (cooling) force. The temperature minimises when the detuning magnitude is half the transition linewidth, $\delta = -\gamma/2$. This is known as the Doppler cooling limit, $T_D = \hbar\gamma/2k_B$, and it is typically on the order of hundreds of microkelvins [61, 65, 76].

2.3.4 Sub-Doppler cooling

In this section, I discuss several sub-Doppler cooling mechanisms, most of which share similar qualitative characteristics. Firstly, I will examine "type-I" transitions, where $F' > F$, and analyse two different combinations of cooling light polarisation: $\text{lin}\perp\text{lin}$ and $\sigma^+ - \sigma^+$ counter-propagating beams. Then I explore "type-II" transitions, where $F' \leq F$ transitions are utilised. These are necessary for the laser cooling of molecules.

Polarization gradient cooling: linearly polarised cooling light

When two counter-propagating beams with orthogonal linear polarisations overlap, they create polarisation gradients. The resulting superposition produces a standing wave with a constant amplitude but varying polarisation at a sub-wavelength scale and is often referred to as the $\text{lin}\perp\text{lin}$ configuration [61, 65]. Let's consider how polarisation varies over one-half wavelength. At the starting point $z = 0$, the polarisation is linear in the y-axis. As we move down the standing wave, the polarisation becomes elliptical until it reaches $z = \lambda/8$, where it becomes right-hand circularly polarised. The polarisation becomes elliptical again until $z = \lambda/4$, where it is linearly polarised in the x-axis. We progress along the standing wave, and the polarisation becomes elliptical until it reaches $z = 3\lambda/8$, where it becomes left-hand circularly polarised. Once again, the polarisation becomes elliptical until it reaches $z = \lambda/2$, and the process starts again, but with the linear polari-

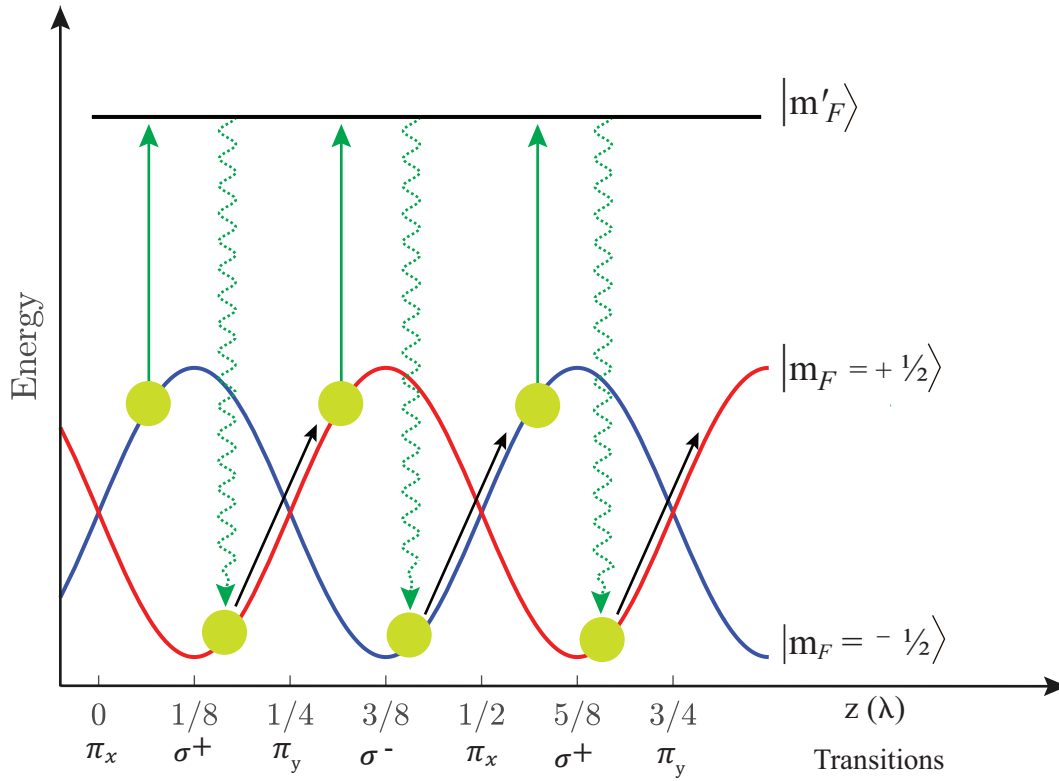


Figure 2.9: Illustration of $\text{lin} \perp \text{lin}$ polarisation gradient cooling for an atom with ground state $F = 1/2$ and excited state $F = 3/2$. The blue and red lines represent the energy levels of the ground state magnetic sublevels $m_F = -1/2$ and $m_F = 1/2$, respectively, which vary due to the AC Stark shift induced by the applied laser light. The circles represent the atom's state, with solid and wavy green arrows indicating optical pumping and radiative decay processes. The black arrows depict the path taken as the system propagates along the z -axis.

sation starting in the opposite y -axis direction [48, 65, 92, 94].

As an atom or molecule moves through the polarisation gradients, the changing polarisation shifts the Zeeman sublevels via the light shift. The ground state energies vary at the sub-wavelength scale due to the light being red-detuned from the transition, favouring $\sigma^\pm$ transitions. Consider an atom with a ground state $F = 1/2$ and excited state $F' = 3/2$, as modelled in the example in figure 2.9 [65]. The AC Stark shifts experienced by ground states are always negative due to the negative detuning. However, the magnitude of these shifts varies depending on the transition intensity. In the case of an atom in the $m_F = -1/2$ state, the σ^- transition is stronger than the σ^+ transitions because the Clebsch-Gordon coefficient is larger. As a result, the $m_F = -1/2$ state undergoes the largest Stark shift when exposed to light with σ^- polarisation. Similarly, the $m_F = +1/2$ state experiences the

greatest shift when illuminated by light with σ^+ polarisation. At $z = \lambda/8$, the atom starts in $m_F = -1/2$, where it has near-maximum potential energy induced by the AC Stark shift. It undergoes a σ^+ transition and then decays to the $m_F = +1/2$ state, which has a minimum AC Stark shift. During decay, the atom loses some energy to the emitted photon due to the difference in AC Stark shifts of its initial and final states. The atom then continues to move through the standing wave, although with a slightly reduced speed. As the atom approaches $z = 3\lambda/8$, it gains potential energy from the AC Stark shift. It undergoes a σ^- transition near $z = 3\lambda/8$ and quickly decays back down to the $m_F = -1/2$ state in which it started. Once again, the atom loses energy, and the cycle continues. Over many cycles, the atom's kinetic energy is stripped away via photon scattering. This process is named after the Greek myth of Sisyphus, who was condemned to push a boulder up a hill only to see it roll back down and had to repeat this task endlessly.

The efficiency of polarisation gradient cooling depends on the atom's velocity. If the atom moves too slowly, it may not reach the maximum of the potential energy curve before getting optically pumped. Conversely, if the atom is moving too quickly, it may reach the downhill side of the curve before being pumped to the excited state, resulting in reduced energy loss per scattered photon. The optimal velocity is achieved when the atom travels a distance of $z = \lambda/4$ in a time $\tau_p = 1/\gamma_p$, where γ_p is the photon scattering rate [65]. In this case, the capture velocity is given by $v_c = \lambda\gamma_p/4$. It is worth noting that this process occurs concurrently with the Doppler cooling force.

Magnetically induced laser cooling: circularly polarised cooling light

Consider a setup where two circularly polarised laser beams, both with the same handedness (σ^+), counter-propagate through an atom with ground state F and excited state $F' = F + 1$. These beams combine to form a standing wave with the same circular polarisation but varying intensity across space. As a result, the atoms in the standing wave experience a spatially varying light shift. This causes them to be optically pumped at the peak light shift and decay to a lower energy m_F state, emitting a scattered photon in the process [65, 95].

However, this form of Sisyphus cooling suffers from an immediate flaw. Since the standing wave has single-handedness, the atoms are optically pumped to the highest m_F state. This imbalance

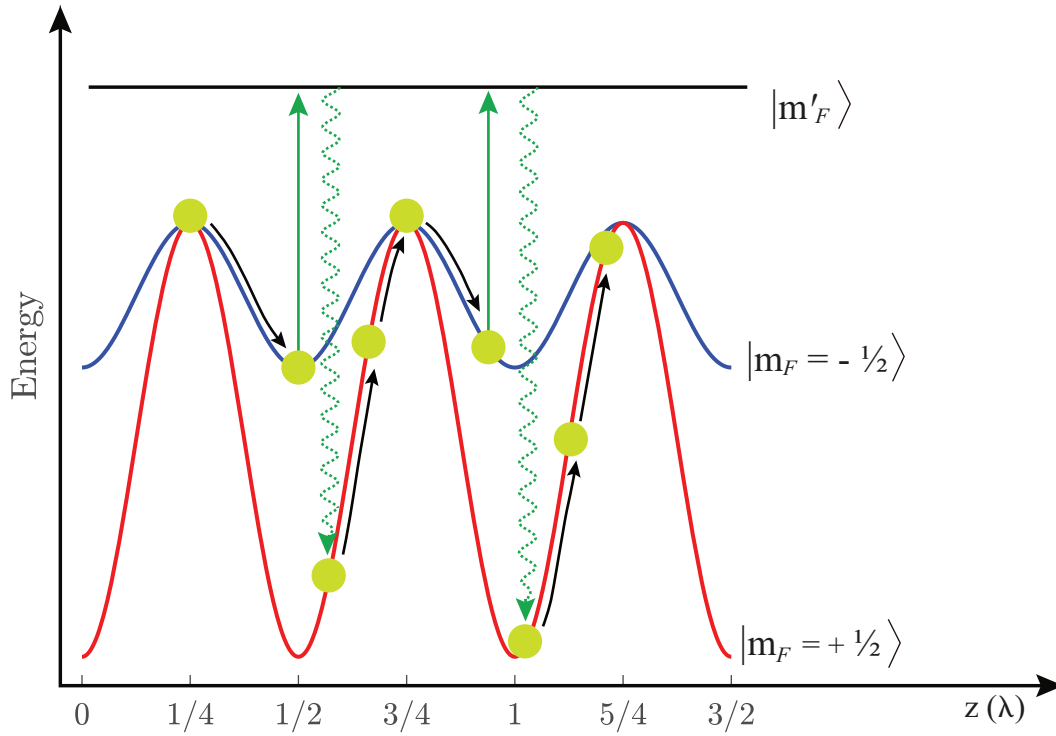


Figure 2.10: Illustration of $\sigma^+ - \sigma^+$ magnetically induced laser cooling for an atom with ground state $F = 1/2$ and excited state $F = 3/2$. The blue and red lines represent the energy levels of the ground state magnetic sublevels $m_F = -1/2$ and $m_F = 1/2$, respectively, which vary due to the AC Stark shift induced by varying intensity of the standing wave. The circles represent the atom's state, with solid and wavy green arrows indicating optical pumping and radiative decay processes. The black arrows depict the path taken as the system propagates along the z -axis.

halts the cycling process and prevents the removal of energy from the system, as the atoms are trapped climbing and descending the same potential hill.

A transverse magnetic field can be applied to the system to address this issue. The applied magnetic field causes Larmor precession of the angular momentum, and by choosing the appropriate magnitude of the field, the precession can be timed such that an atom in sublevel m_F rotates into sublevel $m_F - 1$ at the next node of the standing wave. This rotation helps to prevent the atoms from building up in the higher m_F sublevels, allowing the cooling cycle to continue. Because of the need for a magnetic field, this form of cooling is known as magnetically induced laser cooling.

Figure 2.10 shows this process for an atom with $F = 1/2$ and $F' = 3/2$. Initially, the atom is in the $m_F = -1/2$ state and experiences a light shift that reaches a minimum, where it is optically pumped to the $m_F = 1/2$ state. The atom then climbs a potential hill, and some of its kinetic

energy is converted into potential energy. As the atom climbs, its angular momentum precesses, and when it reaches the next antinode, the atom's magnetic sublevel has changed from $m_F = 1/2$ to $m_F = -1/2$. The cycle then repeats, with the atom losing kinetic energy to scattered photons each time it goes through the cycle [65].

Magnetically induced laser cooling of type II systems

So far, we have focused on "type-I" systems, where the angular momentum of the excited state is greater than that of the ground state, $F' = F + 1$. These systems are advantageous for laser cooling because any polarisation of light can be used to drive transitions, regardless of the ground state sublevel. However, for molecules, we need to consider "type-II" systems, where the excited state can have equal or smaller angular momentum than the ground state, $F' = F$ and $F' = F - 1$ [57, 96]. In such systems, some ground state sublevels cannot be excited by light of certain polarisations, making them "dark" to the cooling light. This poses a challenge to laser cooling.

We can apply a magnetic field to rotate the dark states back into the cooling cycle to overcome this challenge, as previously discussed for type-I transitions. However, unlike in type-I systems, a significant difference arises in that the optimum detuning for a $F' \rightarrow F = F' - 1$ transition is blue-detuned ($\delta > 0$). The light shift for blue-detuned light is opposite to the shift caused by red-detuned light, with the ground and excited state energies shifting closer together [65]. This configuration results in Doppler heating of atoms/molecules with higher velocities while slower-moving ones experience the dominating Sisyphus force, which slows them [57, 93, 96–99].

Consider a type-II transition with $F' = 0$ and $F = 1$, where the only excitation possible with linearly polarised light is the $m_F = 0$ to $m_{F'} = 0$ transition. Figure 2.11 illustrates the cooling process of this system, with the bright state $m_F = 0$ and dark states $m_F = \pm 1$ shown separately. The system starts in the bright state and, at the top of the potential hill, gets optically pumped into one of the dark states. The applied magnetic field rotates the system's angular momentum as it passes through the wave node, and the system rotates from the dark state into a bright state. Another potential hill is then climbed, and the cycle repeats, converting kinetic energy to potential energy and removing it by scattering photons. This is the laser cooling mechanism used in the work described in this

thesis.

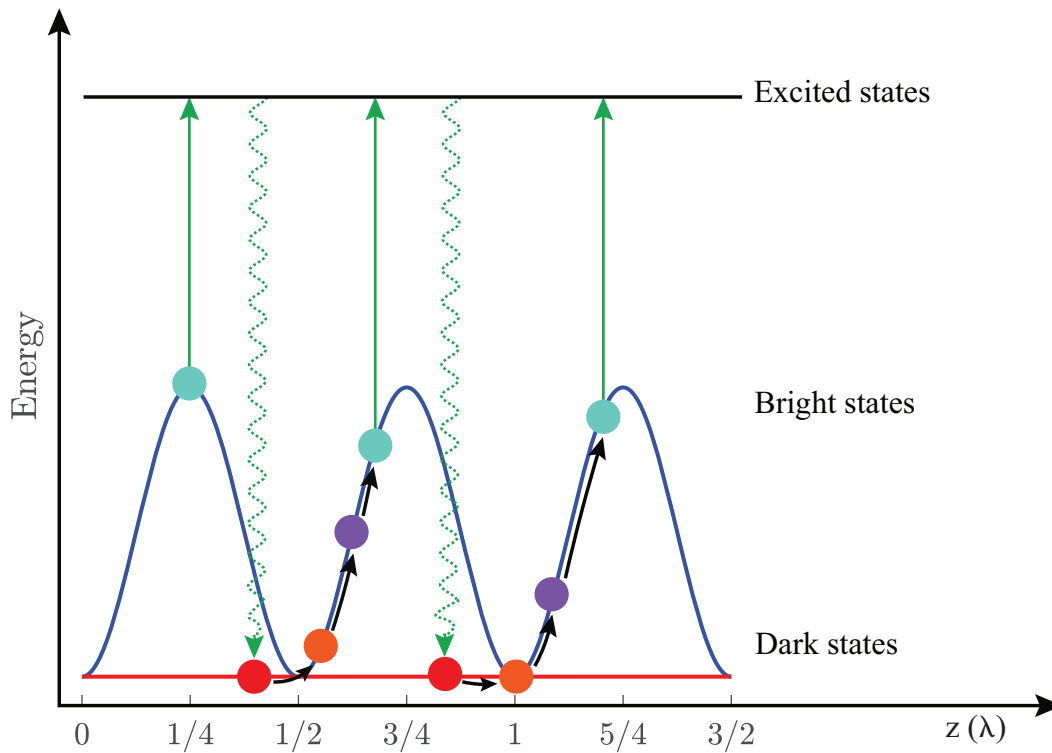


Figure 2.11: Illustration of magnetically induced laser cooling for a type-II transition. The blue and red lines represent the energy levels of the bright and dark states, respectively. The energy levels of the bright state vary due to the AC Stark shift induced by the intensity-varying standing wave. The circles represent the system's state, with solid and wavy green arrows indicating optical pumping and radiative decay processes. The black arrows depict the path taken as the system propagates along the z -axis.

Laser cooling of atoms or molecules using blue-detuned light results in a velocity-dependent force that exhibits deceleration at low velocities and acceleration at high velocities. This behaviour arises from the combined effects of Sisyphus cooling and Doppler heating. The force curve reaches a zero crossing at a specific velocity, known as the critical velocity. At velocities greater than the critical velocity, the atoms or molecules are accelerated, while they are decelerated at velocities below the critical velocity. Understanding the concept of critical velocity is important for interpreting the laser cooling results presented in chapter 4.

Chapter 3

Experimental methods

This chapter presents a detailed account of the experimental methods employed to measure the electric dipole moment of the electron, comprising several components that are described in detail. The chapter begins by providing an overview of the experimental setup in section 3.1, which includes a comprehensive description of the vacuum systems that have been assembled and are critical for the success of the experiment. Section 3.1.2 discusses these systems in detail. Then, to provide a more in-depth understanding of the experiment, the chapter focuses on five key systems: the molecular source (section 3.2), the laser systems used for laser cooling, optical pumping, and detection (section 3.3), the microwave systems used for optical pumping (section 3.4), and the computer control systems (section 3.5).

3.1 Overview of experiment setup

3.1.1 The beamline

An apparatus drawing is presented in figure 3.1, while figure 3.2 displays a cutaway view. The apparatus comprises a series of vacuum chambers aligned horizontally, creating a beamline for the molecular beam to travel through. The molecules travel through the chambers in the specified sequence:

1. A cryogenic buffer gas source which produces a pulsed beam of molecules
2. A hexapole magnetic lens
3. Transverse laser cooling
4. Optical pumping
5. Superposition preparation
6. The eEDM interaction region
7. Superposition recombination
8. State selective molecular detectors

The cryogenic buffer gas source is located in region (1) of figures 3.1 and 3.2, and it's discussed in detail in section 3.2. This source generates a pulsed beam of YbF molecules with forward speeds as low as 150 ms^{-1} . The molecules produced by this source are cold, have relatively low divergence, and primarily populate the low rotational states $N = 0, 1$, and 2 .

After leaving the molecular source, the beam passes through a magnetic lens located in region (2), as depicted in figures 3.1 and 3.2. Although it has not yet been integrated into the beamline, the lens has been constructed and is awaiting installation. Its function is to focus the molecular beam so that more molecules are laser-cooled. The magnetic lens is extensively discussed in chapter 5.

In region (3) of figures 3.1 and 3.2, the molecules travel through an optical molasses and undergo two-dimensional transverse laser cooling. This is the topic of chapter 4 and is the second part of the "molecular funnel" discussed in chapter 5. By performing transverse laser cooling, we collimate the molecular beam. As such, the coherence time of the molecules is greatly improved and, in turn, so is the sensitivity of an eEDM measurement.

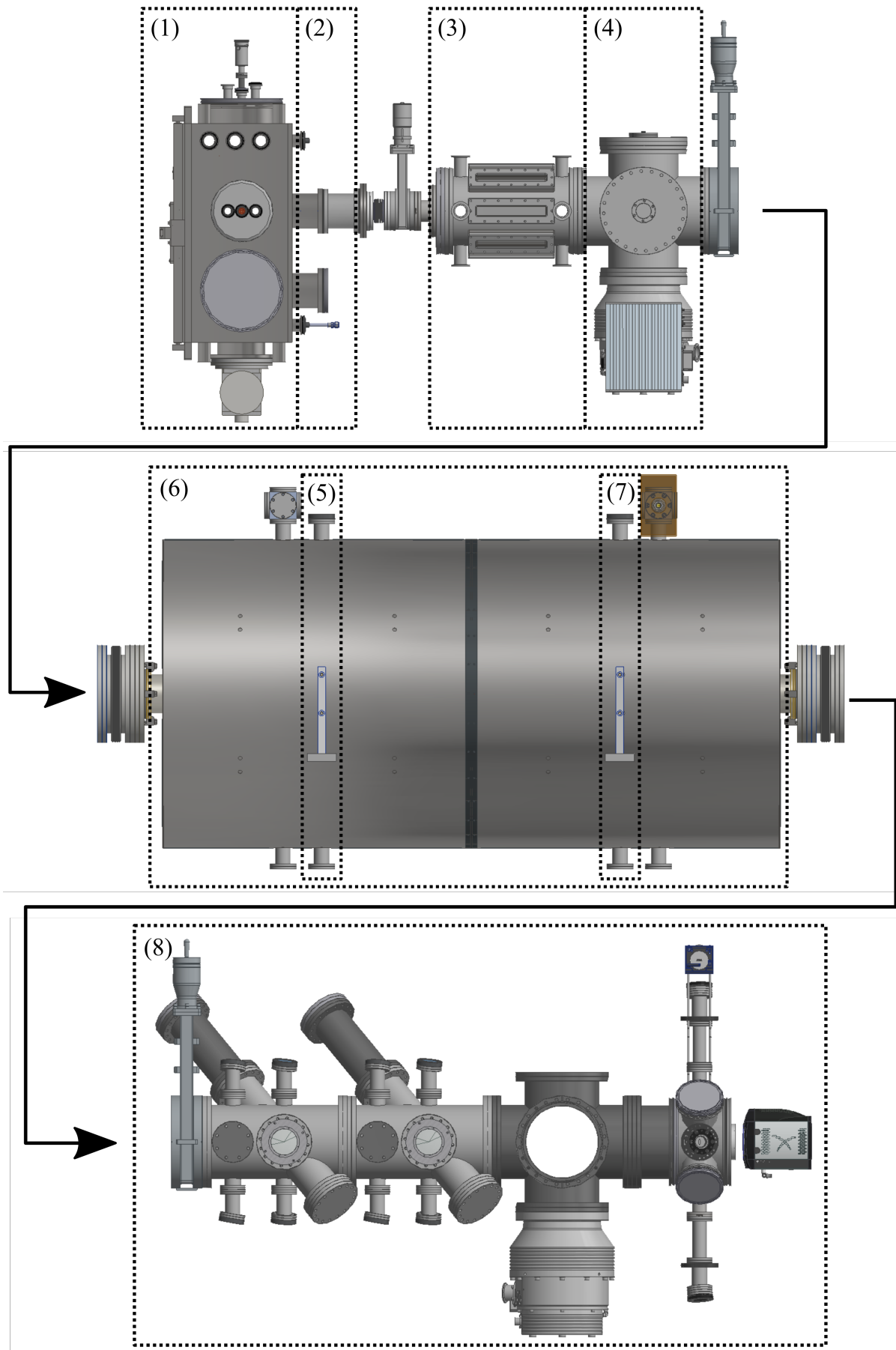


Figure 3.1: CAD drawing of the beamline: (1) molecular source, (2) magnetic lens, (3) two-dimensional transverse laser cooling, (4) optical pumping, (5) eEDM state preparation, (6) eEDM spin precession measurement region, (7) eEDM state recombination, (8) state-selective detection.

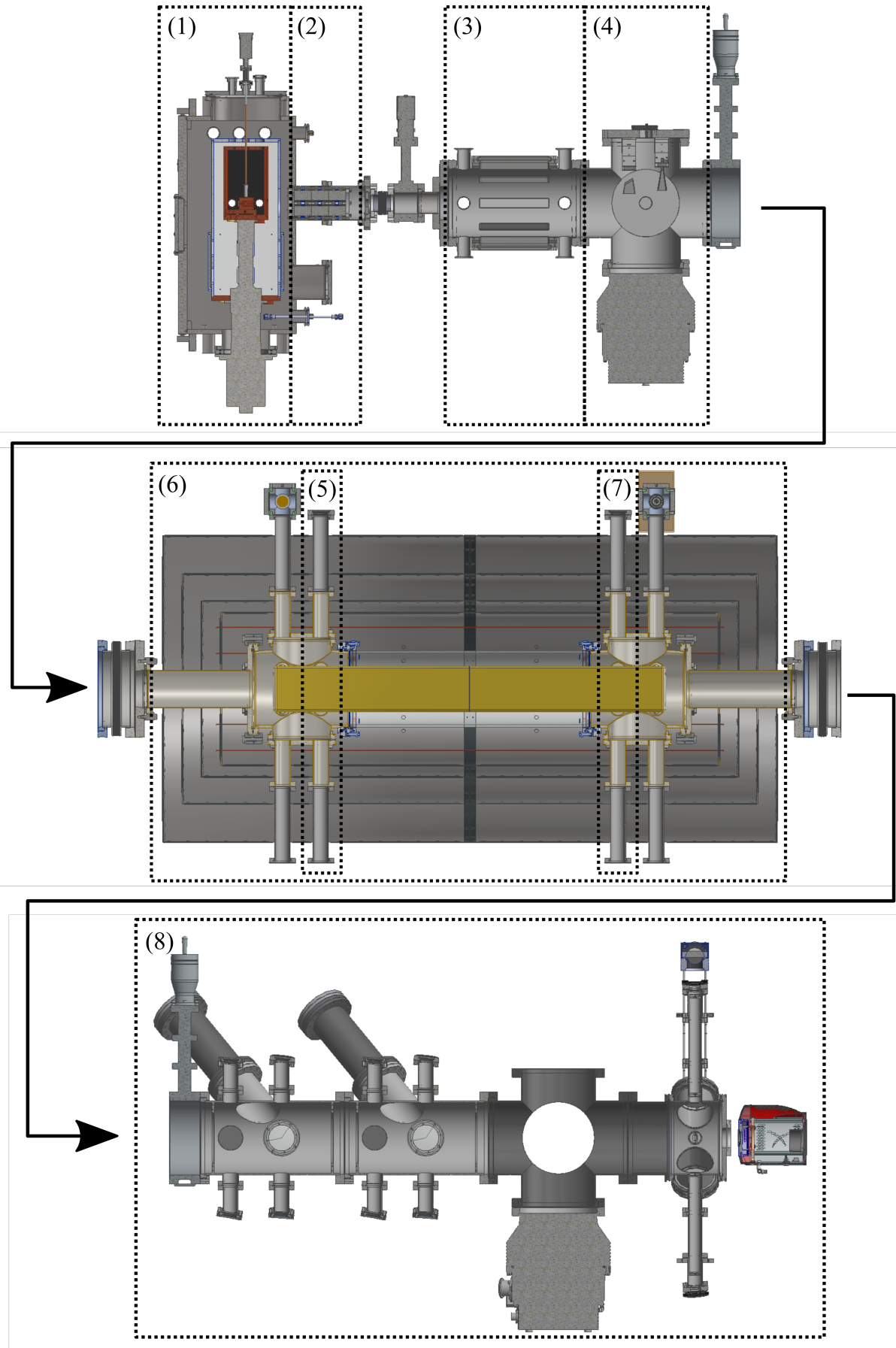


Figure 3.2: Half-section view of the beamline CAD drawing: (1) molecular source, (2) magnetic lens, (3) two-dimensional transverse laser cooling, (4) optical pumping, (5) eEDM state preparation, (6) eEDM spin precession measurement region, (7) eEDM state recombination, (8) state-selective detection.

The molecular state suitable for laser cooling, $N = 1$, differs from the one required for measuring the eEDM, $N = 0$. Therefore, in region (4) of figures 3.1 and 3.2, the molecules need to be transferred to their ground rotational state. This is accomplished by applying microwaves to couple the first and second excited rotational states while simultaneously using laser light to pump the molecules from their second excited rotational state to their ground rotational state. Additionally, laser light is used to pump molecules out of the $(N = 0, F = 1)$ state to ensure that all molecules are in the $(N = 0, F = 0)$ state. The details of this process are explained in chapter 6.

Once the YbF molecules are in the $(N = 0, F = 0)$ state, the next step is to prepare them for eEDM measurement. This will be done using stimulated Raman adiabatic passage (STIRAP) in region (5) of the apparatus shown in figures 3.1 and 3.2. This technique involves using two partially overlapped optical laser beams to coherently transfer the molecule population from the $F = 0$ state to the superposition state $\frac{1}{\sqrt{2}}(|F = 1, m_F = +1\rangle + |F = 1, m_F = -1\rangle)$.

The molecules then travel through the eEDM interaction region, which is located in region (6) of figures 3.1 and 3.2. Here, two parallel plates with a high voltage applied produce a uniform electric field that interacts with the molecules. The plates have a length of 1 m and are separated by a gap of 1 cm. A weak magnetic field is also applied by coils outside the vacuum chamber, causing spin precession and imparting a phase on the superposition state. Careful attention has been given to ensure the electric field plates are flat and smooth, creating a uniform field between them. Designing the supporting electronics for the plates is also critical. High voltage requirements can result in discharges between wires and the chamber or between the plates, adversely affecting the eEDM measurement. Four nested magnetic shields surround the interaction region to reduce measurement noise caused by background fields.

Region (7) of figures 3.1 and 3.2 is dedicated to the recovery of the population from the superposition state. This will be achieved through a reverse process of the STIRAP combination, where molecules in the $\frac{1}{\sqrt{2}}(|F = 1, m_F = +1\rangle + |F = 1, m_F = -1\rangle)$ state are transferred back to the $N = 0, F = 0$ state, while molecules in the $\frac{1}{\sqrt{2}}(|F = 1, m_F = +1\rangle - |F = 1, m_F = -1\rangle)$ state remain in $N = 0, F = 1$. The separation of these two populations enables independent detection.

Finally, in region (8) of figures 3.1 and 3.2, the molecules enter the state-selective detectors. There

are two spatially separated detectors for eEDM measurements, one for the $N = 0$, $F = 0$ state and the other for the $N = 0$, $F = 1$ state. Inside each detector's vacuum chamber, a microwave horn is mounted at a 45-degree angle to couple the molecules to the first excited rotational state. The molecules are then optically pumped with P(1) light until they are all in higher vibrational states. Optical detection is performed using both PMTs and cameras, each providing unique diagnostic information. PMTs operate on a shorter time scale and can therefore gate the signal in time, while cameras offer better spatial resolution. At the end of the beamline, there is an additional detector facing the molecules in the direction of their propagation. This detector is essential for setting up laser cooling in both transverse axes. Further details about this final detector can be found in chapter 4, where laser cooling is discussed.

3.1.2 Vacuum systems

The illustrations in figures 3.1 and 3.2 show a series of horizontally aligned vacuum chambers that comprise the beamline. The rough pumping is performed by three scroll pumps, which then function as backing pumps for the three turbomolecular pumps, enabling pumping to ultra-high vacuum. The turbomolecular pumps are located below the beamline in the optical pumping region and detection region, while the third pump is mounted on the side of the source chamber (not visible in the drawings as it is behind the chamber). The beamline is typically operated at a pressure of around 5×10^{-7} mbar, measured using pressure gauges installed in the source, interaction region, and detection region. It is worth noting that the pressure in the source may be higher during operation due to the continuous flow of buffer gas.

To facilitate maintenance, some critical segments of the beamline are separated by bellows and gate valves. This is particularly significant because it enables individual sections of the beamline to be opened to the atmosphere without requiring the whole system to be vented. This allows maintenance work to be performed without having to bake down the entire beamline to low pressures, which speeds up maintenance procedures.

3.2 Molecular source

In the words of J. Hudson, "the starting point of any day in the lab is to make a beam of molecules" [9]. Due to its high reactivity, YbF cannot be readily purchased and must be produced as needed. This section explores the different types of molecular sources that can be utilised for this task, followed by a detailed description of our chosen design: a cryogenic buffer gas source.

3.2.1 Types of molecular sources

Our research group at Imperial College has been actively developing molecular sources for many years and continues to do so. There are three distinct methods for creating YbF molecules: effusive, supersonic, and buffer gas sources, as detailed in [9, 43, 48]. Figure 3.3 shows that these sources produce molecular beams with significantly different velocity distributions, resulting in different molecule rotational states. As our eEDM measurement and laser cooling processes use molecules in the $N = 0$ and $N = 1$ states, it is desirable to utilise a colder molecular beam.

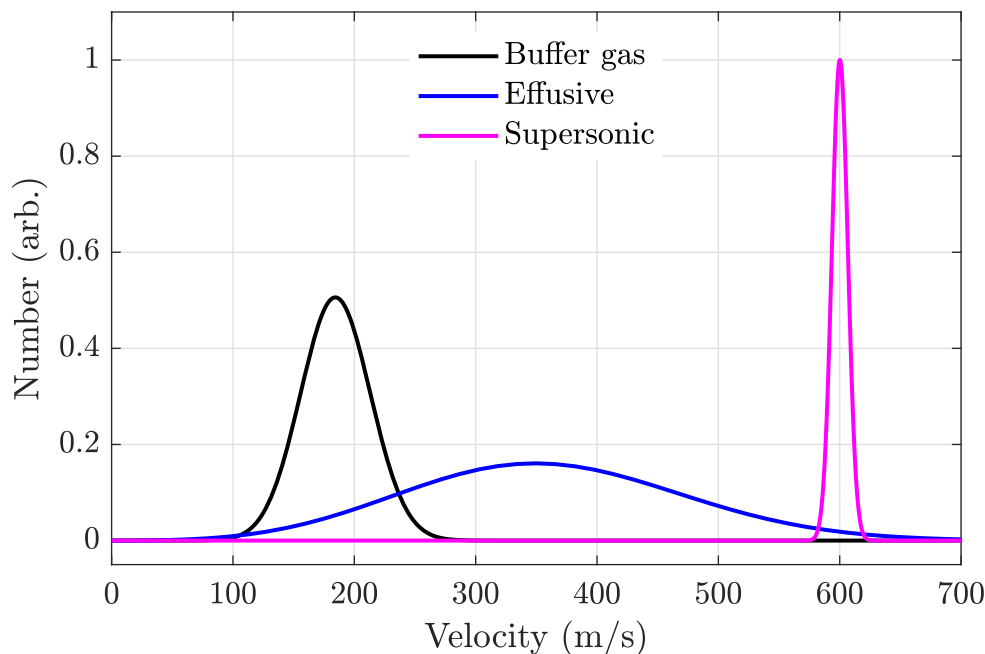


Figure 3.3: Simulated probability distributions functions for the three types of YbF molecular sources.

The effusive source is a method of creating YbF molecules by heating precursor compounds. How-

ever, due to the high temperatures required, the resulting molecular beam is also hot, and the molecules have a broad range of velocities, as illustrated in figure 3.3. The high divergence of the beam makes it challenging to obtain a precise measurement of the eEDM as it detrimentally affects the coherence time. Although it was the first source attempted, its limitations have prompted the exploration of other methods. J. Hudson's thesis provides further details on this source [9].

A previous eEDM experiment at Imperial College used a supersonic YbF source created by ablating a Yb metal target with a Nd:YAG pulsed laser in the presence of SF₆ gas [2, 3, 41, 100]. The ablated Yb reacts chemically with the SF₆ gas to create YbF gas, which then cools as it expands supersonically from the high-pressure environment where it is generated to the low-pressure chamber where the eEDM measurement is conducted. With a mean velocity of approximately 600 m s^{-1} , the supersonic beam is much faster than that produced by the effusive source [3]. Although it diverges less than the effusive beam after passing through a skimmer, the high forward velocity limits the time the molecules spend in the interaction region, thereby reducing the eEDM sensitivity. Our group has investigated supersonic sources in detail, and more information can be found in several theses [2, 3, 41, 100].

Buffer gas sources function similarly to the previously mentioned sources in which Yb is reacted with a fluorine donor. However, the main difference is that this reaction occurs in the presence of a cold, inert gas, known as a "buffer gas", typically a noble gas [43, 48, 63]. The resulting YbF molecules are then cooled by elastic collisions with the buffer gas atoms in a cryogenically cooled container. Once the molecules exit through a hole in the container wall, they expand and cool further. Compared to the supersonic and effusive sources, this source type produces a much slower and colder beam with significantly lower divergence, making it an ideal candidate for high-precision measurements of the eEDM.

3.2.2 The YbF buffer gas cooled source

In several experiments, the group uses cryogenic buffer gas sources to make beams of CaF and YbF molecules. These sources are all variants of the same design [101]. For a comprehensive review of

buffer-gas cooled beams, refer to Hutzler et al. (2012) [43].

In this section, I provide a detailed description of our YbF molecular source, starting with the "cell" where the molecules are created and then moving on to the infrastructure that supports the operation of our source.

The cell

Our YbF molecular source features a copper cube that sits atop a Sumitomo SRDK-415 cryocooler, with a cylindrical chamber inside the cube where YbF molecules are produced. This part of the source is called "the cell". All copper components in the source are machined from oxygen-free copper to enhance thermal conductivity and minimise volatility under high vacuum conditions. Depending on thermal loads, the cryocooler can cool the cell to a temperature range of 4 – 5K. Figure 3.4 includes three different views of the cell, which depict the fully assembled cell, an exploded view of the cell from a different angle, and a half-section view from above.

The pulsed Nd:YAG laser (Continuum, Minilite II) generates light that enters the cell through a glass window mounted on the end of a copper tube called the "snorkel." The laser ablates a solid Yb target, producing a vapour of Yb as well as Yb dust that coats the internal surfaces of the cell. To reduce the amount of dust deposited on the window, the snorkel increases the separation between the window and the Yb metal target. The laser produces pulses of ~ 10 mJ with a duration of 10 ns at a repetition rate of 5 Hz. Previous designs used a laser focused on the Yb rod, which resulted in the drilling of holes into the rod after several thousand pulses¹. However, we found that careful tuning of the laser power and removal of focusing optics prevented this issue in the current design. Instead, the Yb rod surface becomes blackened over many ablation pulses, eventually decreasing the yield significantly. When this happens, the Yb rod can be rotated and raised/lowered using a threaded copper rod mounted on the end of the rod, which changes the position that the Nd:YAG light is incident upon, reducing the need for time-consuming maintenance.

¹Whilst I worked with the previous source, I've not included a description here as it doesn't pertain to the current beamline, and there are only minor differences. A description of the cell we used previously can be found in M. Trigatzis' thesis [48].

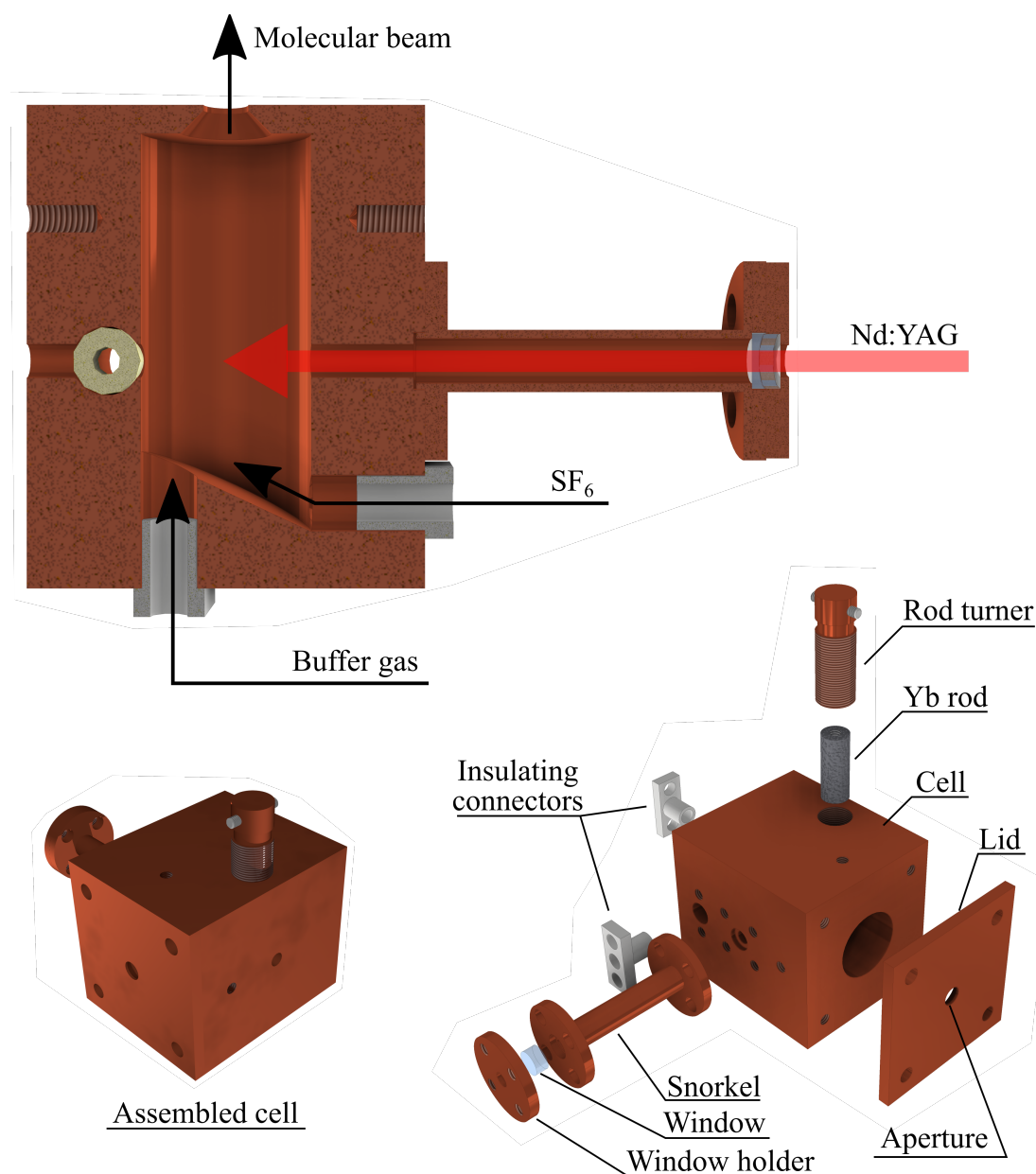


Figure 3.4: CAD drawings of the cell used in the experiment. The bottom left view displays the fully assembled cell, while the bottom right view provides an exploded perspective with labels of the individual components. The top view presents a half section of the cell from above. Laser pulses from the Nd:YAG laser are introduced to the cell via a window on the right, while SF₆ and buffer gases enter from the bottom right and bottom left corners, respectively. YbF molecules exit the cell through an aperture located at the top of the drawing.

SF₆ gas flows continuously into the cell at a flow rate of 0.005 SCCM and chemically reacts with the hot Yb vapour to create YbF molecules. However, if too much SF₆ (0.1 SCCM) flows into the cell, the molecular output drops to almost nothing, causing the source to be "poisoned", and flux output can only be restored by cleaning the cell. Cleaning the cell reveals a light grey powdery substance in the bore, similar to a dust bunny. The freezing temperature of SF₆ is 222 K, so the

pipe carrying the SF₆ gas to the cell is both insulated from the cryogenically cooled cell using a plastic connector and heated using a resistive heating element to prevent the gas from freezing before it reaches the cell.

Cryogenically cooled helium gas flows continuously into the cell at a rate of 1 SCCM, where it thermalises with the hot YbF molecules through elastic collisions. The gas travels through a pipe wound around a copper pipe, forming a "bobbin" mounted on the cold head of the cryocooler, ensuring that the helium is cooled to cryogenic temperatures before it reaches the cell.

Finally, the He and YbF gas mixture exits the cell via an aperture. The aperture is part of a copper plate mounted on the front of the cell. On the side of the plate facing the cell, the aperture is bevelled.

Supporting infrastructure

Figure 3.5 illustrates a half-section drawing of the molecular source, which is housed in a cuboid vacuum chamber with a door for easy maintenance access. Vacuum feedthroughs located along the top and bottom sides of the chamber enable sensors and other devices to be connected. The background pressure in the source chamber is measured by a combined Pirani Bayard-Alpert pressure gauge that can detect a range of pressures, from atmospheric pressure to as low as 5×10^{-10} mbar.

The cryocooler, which is mounted vertically at the bottom of the chamber, consists of two stages. The first stage achieves a temperature of ~ 31 K, whereas the second stage, on which the cell is mounted, reaches a temperature range of 3.5 – 5 K depending on the thermal load. A large aluminium box is placed on the first stage, which we refer to as the "radiation shield" because it encompasses the second colder stage and protects the contents from black body radiation emitted from the environment. This radiation would otherwise increase the thermal load on the cryocooler significantly. Additionally, a bobbin, where the buffer gas is pre-cooled, is mounted on the first stage before moving to the second-stage bobbin. Although the bobbins are not visible in Figure 3.5, the second-stage bobbin is shown in Figure 3.6.

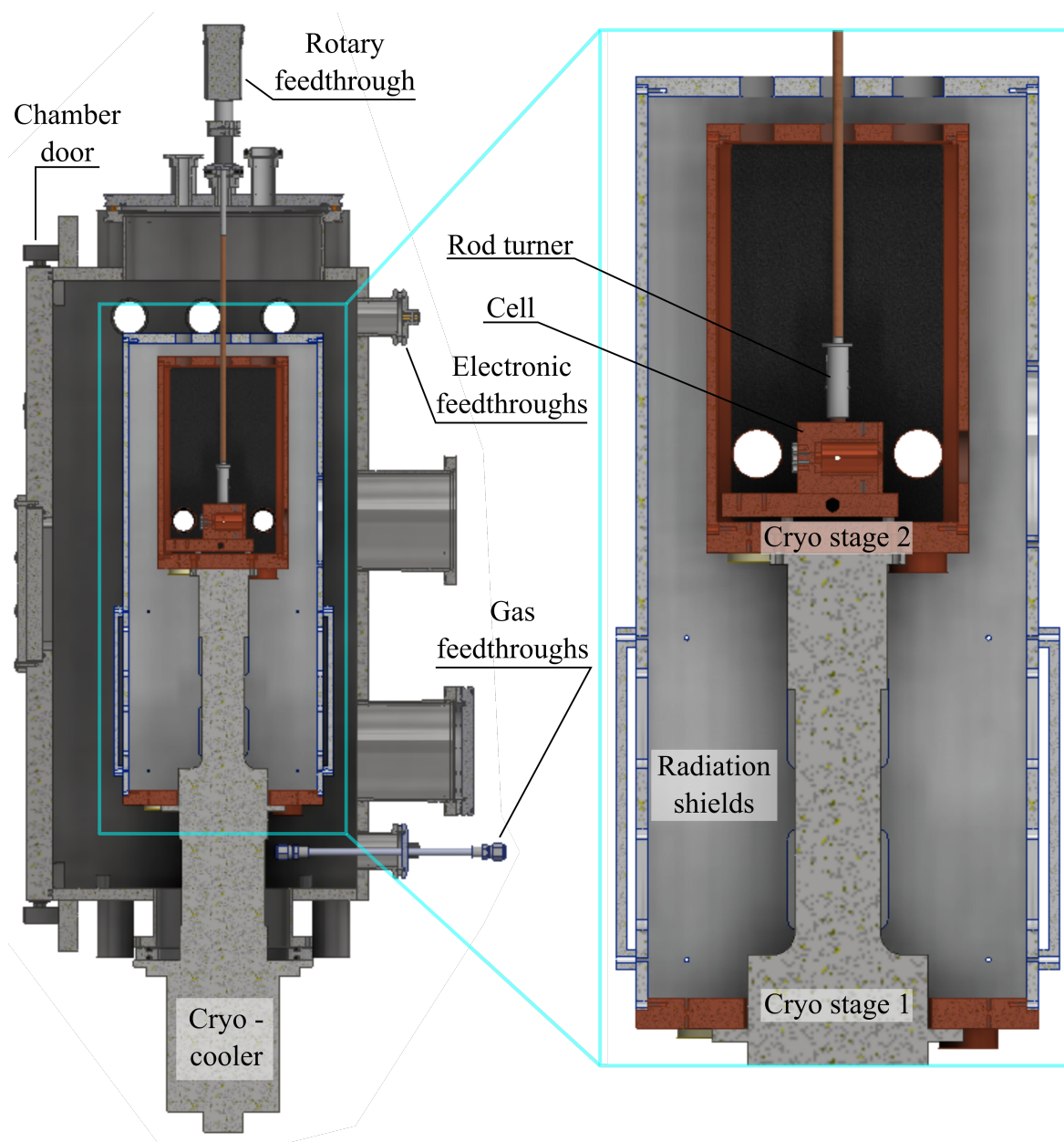


Figure 3.5: Half-section CAD drawing of the source, displaying the internal components of the first and second stages of the cryocooler and the mounted components. The internal gas pipes and bobbins are not visible as they are out of the page due to the half-section cut. The zoomed-in view on the right shows a detailed view of the first and second stages of the cryocooler.

A long plastic rod is used to connect the Yb rod turner to the vacuum rotary feedthrough. We initially experimented with stainless steel, which has a relatively poor thermal conductivity compared to other metals. However, we found that this material increased the thermal load on the second stage of the cryocooler excessively.

Figure 3.6 presents a detailed drawing of the second-stage components of the molecular source.

The core of the setup is the cell, which is mounted on a rectangular copper plate that sits on top of a square copper baseplate. Surrounding the cell is a copper box, which features openings for the molecular beam, laser light, detection light, and Yb rod turner. The box also includes additional holes, which were used for testing different cell positions during the design phase but are now sealed with threaded metal disks.

Since the SF₆ gas pipe must be insulated from the cryogenically cooled surfaces, a nylon spacer attached to the baseplate prevents accidental contact with the cold baseplate. The surfaces of the copper box are coated with a thin layer of activated charcoal, which is attached to the copper surfaces using a thin coat of Stycast 1266 epoxy. When cooled to cryogenic temperatures, the activated charcoal acts as a sorption pump for helium. This is crucial because turbomolecular pumps have poor helium pumping efficiency. Without sorption pumps, the vacuum pressure of the beamline quickly increases to a level where the molecular beam cannot freely propagate down the line.

If the source runs for too long, the charcoal will become saturated, and the background pressure of the source chamber will increase. In this scenario, the cold surfaces could become thermally connected to the room temperature environment, causing a runaway reaction in which the charcoal quickly warms and the helium desorbs, further improving the thermal connection to the environment. Additionally, the cold surfaces cryopump other background gases in the chamber, which would quickly sublime and increase the background pressure. To avoid this, the source is refreshed daily by warming it to 330 K overnight and then cooling it down again for the next day's work. This process, known as a "refresh," has been automated for convenience. More details about the refresh automation software are discussed in section 3.5.3.

Several temperature sensors and heaters are distributed throughout the source. Lakeshore DT670 silicon diodes facilitate temperature measurements on the first and second cryocooler stages, as well as the SF₆ gas pipe, buffer gas pipe, and cell. This diagnostic information is essential for both the reliable production of YbF molecules and the safe refreshing of the source. In addition, heaters are installed on the cell, SF₆ gas pipe, buffer gas pipe, and the first and second stages of the cryocooler. The heaters on the gas pipes provide control of the gas temperatures, while the other heaters facilitate a faster and better-controlled refresh of the source.

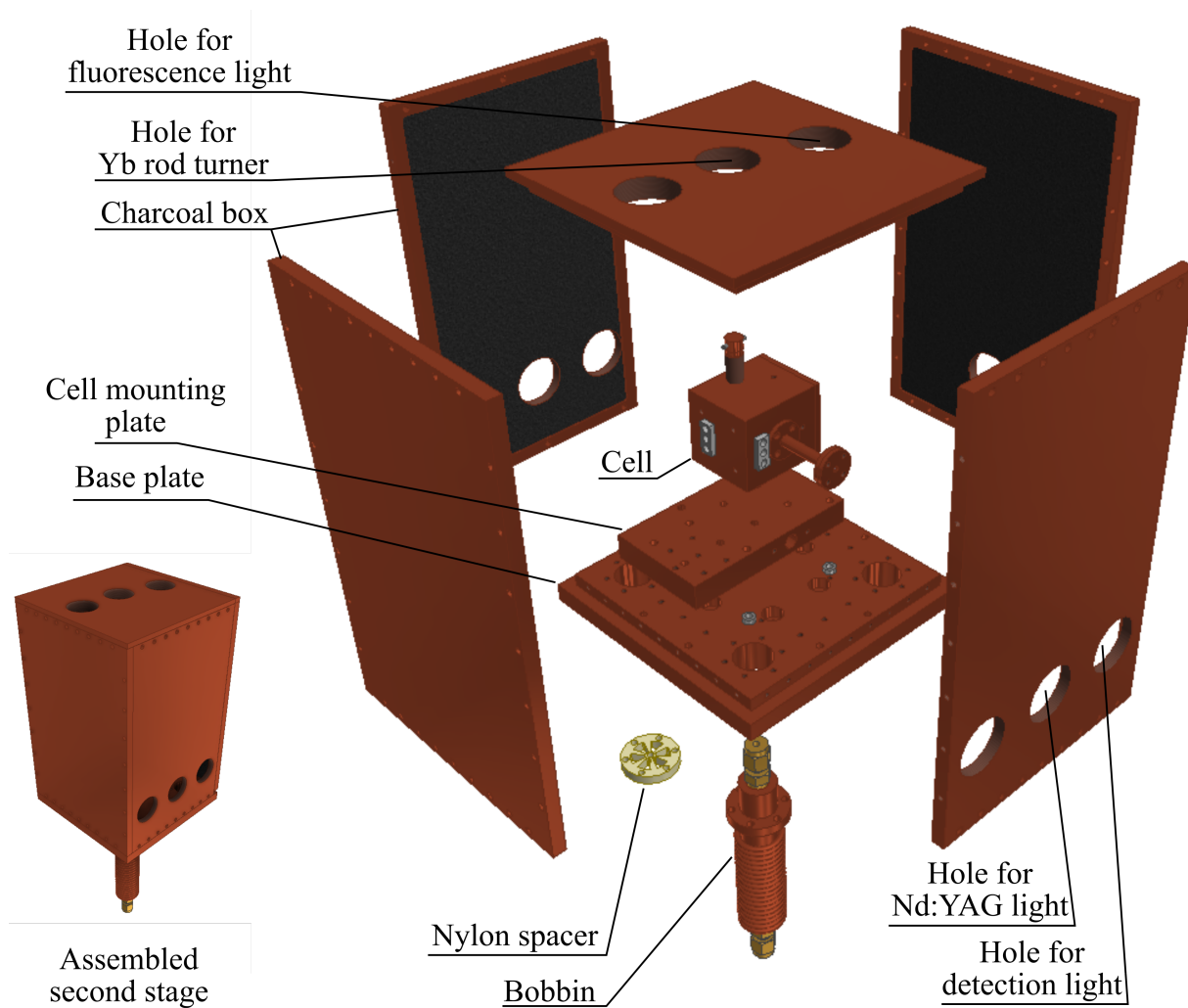


Figure 3.6: CAD drawing of the components on the cryocooler second stage. In the bottom left is an assembled view. Beside it is an exploded view of the components. The plastic Yb rod turner and gas lines are not shown. Also not shown are the circular windows for the optical and infrared light to enter/exit the box.

3.3 Laser systems

The laser systems required for this experiment are numerous and complex. The following sections describe the specifics of the laser systems that have already been built and are currently used. Section 3.3.1 outlines the laser systems needed for cooling and repumping the molecules, totalling three lasers. Section 3.3.2 discusses a single laser used for light-induced fluorescence measurements of the molecules. Two lasers are required for optical pumping of the molecules, detailed in section 3.3.3. Finally, section 3.3.4 describes the frequency tuning and locking apparatus.

In general, each laser system requires multiple additional frequencies to address the hyperfine structure of the molecules. This is achieved through the use of acousto-optical modulators (AOMs) and electro-optical modulators (EOMs). The individual requirements of each laser system are discussed in detail.

It should be noted that the laser systems required for STIRAP and eEDM state selective detection, which may potentially make use of some of the existing laser systems, are not yet complete and are therefore not discussed here.

3.3.1 Cooling and vibrational repump lasers

When molecules transition from the cooling state $X^2\Sigma^+ (\nu = 0, N = 1)$ to the excited state $A^2\Pi_{1/2} (\nu' = 0, J' = 1/2)$, they tend to scatter about 14 photons on average before decaying to a higher vibrational state of X . To bring back these molecules to the cooling cycle, we focus on exciting the first and second vibrational states of X , but not higher ones. For this, we utilize three laser systems for cooling and repumping, with their transitions' frequencies and branching ratios detailed in section 2.1.6. More information about these laser systems can be found in M. Trigatzis' thesis [48].

All three laser systems rely on a similar approach for generating the necessary green light. Unfortunately, laser diodes at these frequencies are currently unavailable in the market. As a result, we use infrared laser diodes to generate the light, which is then power amplified and frequency doubled to produce the green laser light.

The necessary light for the cooling transition is generated using a Toptica DL Pro 021187 that produces 28 mW of 1104 nm seed light. This is then power amplified and frequency doubled using an MPB Communications visible Raman fibre amplifier (VFRA) and its integrated second harmonic generation (SHG) module. The resulting output is approximately 1.7 W of 552 nm laser light.

A separate laser system is used to repump the first excited vibrational state back into the cooling transition via the $A^2\Pi_{1/2} (\nu' = 0, J' = 1/2)$ state. The infrared seed light is generated using a Toptica DL100 Pro 17430 that produces 34 mW of 1136 nm laser light. This is then power amplified and

frequency doubled by another MPB Communications VFRA and SHG system, producing an output of 900 mW of 568 nm laser light.

To repump the second excited vibrational state back into the cooling transition via the [557] state, a separate laser system is used. Seed laser light is generated using a Toptica DL100 L_03098 that produces 34.5 mW of 1130 nm laser light. It is then power amplified to 160 mW using a home-built tapered amplifier that utilises a Toptica TA-1135-0500-1 chip. The resulting amplified light is coupled into a fibre and passed through a waveguide PPLN frequency doubler (NTT Electronics WH-0565-000-F-B-C). Although the best output achieved is 60 mW of 565 nm laser light [48], the system usually produces 45-50mW due to thermal drifts in the amplifier.

Figure 3.7 depicts the optics circuit after generating the green light. The three lasers enter the circuit at the figure's top and left sides, and the light exits through several fibres that transport it to various parts of the experiment. AOMs and EOMs are represented by red and purple boxes, respectively, with their modulation frequencies written inside.

The cooling light is injected into the optics circuit from the left side of figure 3.7. It is split into three beams using half-wave plates and polarising beam splitters (PBSs). The back reflection from the first half-wave plate is coupled into a fibre and sent to a cavity for frequency tuning and locking. Two of the beams are frequency-shifted by 159 MHz and 192 MHz using AOMs. The three beams are then recombined (with the repump lasers) using half-wave plates and PBSs. A small amount of the light is taken to a wavemeter to monitor the frequency. The recombination process results in two beams: one for each of the transverse axes to be cooled. Polarization-maintaining (PM) optical fibres are then used to transport the beams to the beamline.

From the top left, the vibrational repump lasers enter figure 3.7. A beam sampler ($\sim 3\%$ reflectance) is used to immediately sample some light from the 568 nm laser and send it, via an optical fibre, to a cavity for laser tuning/locking (see section 3.3.4). AOMs and EOMs are used to create the frequencies needed to address hyperfine structure. Then, the three resulting beams are combined using a dichroic mirror and PBS. Some of the light exiting the PBS is sent to the "cleanup region," where it is used to repump molecules after laser cooling. Most of the light exits the other face of the PBS and is combined with the 552 nm cooling light using a dichroic mirror. A small amount

of repump light is reflected from the imperfect dichroic mirror; this is sent to the wavemeter for frequency monitoring.

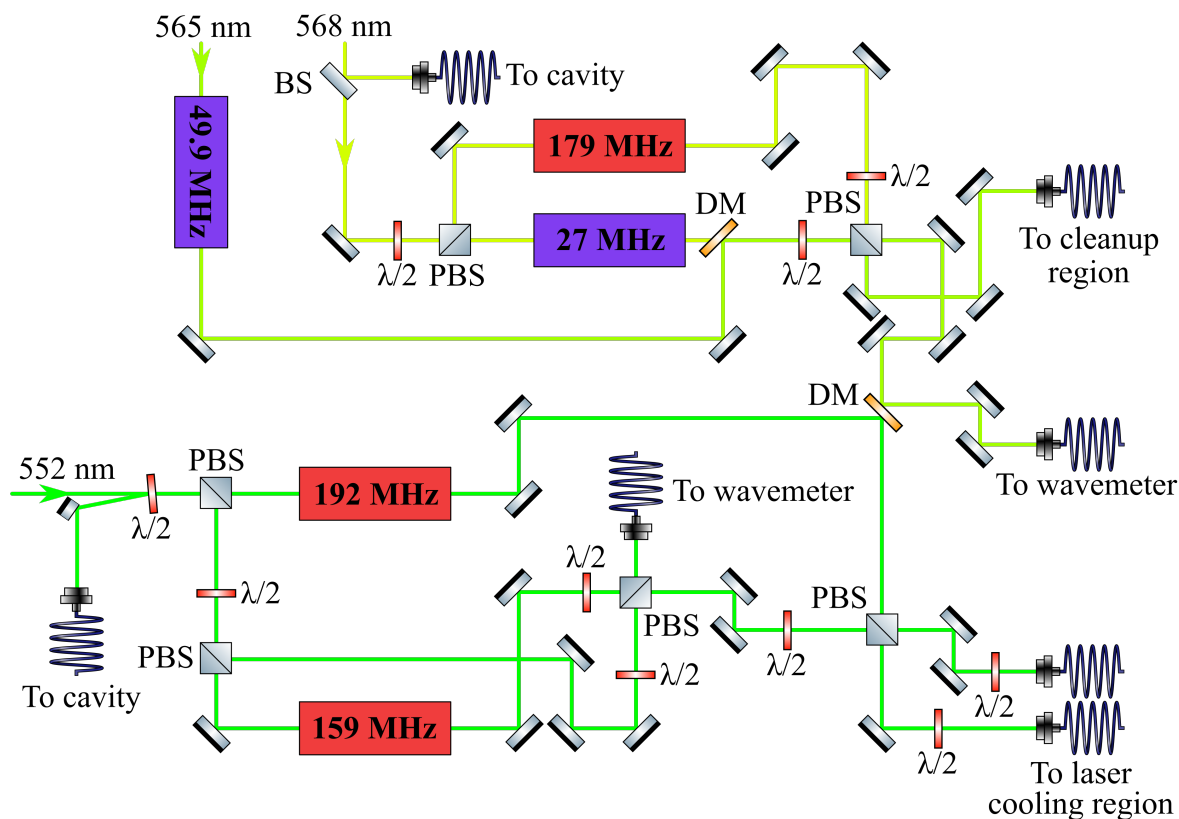


Figure 3.7: Diagram illustrating the cooling and vibrational repump lasers circuit. The components are labelled as follows: BS for a beam sampler, PBS for polarising beam splitter, $\lambda/2$ for the half-wave plate, and DM for a dichroic mirror. The red and purple boxes represent AOMs and EOMs, respectively, with their modulation frequencies written inside. At the end of the optical circuit, several fibres transport the laser light to different parts of the experiment.

3.3.2 Probe laser

The detection of molecules is achieved through light-induced fluorescence, where, in the laser cooling experiments, the $X^2\Sigma^+ (\nu = 0, N = 1) \rightarrow A^2\Pi_{1/2} (\nu' = 0, J' = 1/2)$ transition was utilised. This transition is advantageous because it allows for calculating the number of detected molecules due to the well-defined average number of photons scattered before the molecules become dark to the probe light.

The probe laser begins as an infrared laser produced by a homebuilt extended-cavity diode laser

(ECDL) [94], which is then fibre-coupled into a Keopsys ytterbium fibre amplifier (YFA) to produce approximately 12 mW of 1104 nm seed light. The amplified IR laser light is frequency doubled using a free-space periodically-poled lithium niobate (PPLN) crystal, resulting in up to 500 mW of 552 nm laser light.

The probe laser optics circuit is shown in figure 3.8. The 552 nm light enters from the left-hand side and is split into three beams, with two of the beams being shifted in frequency using AOMs in the same manner as the cooling light. Recombination of the beams is accomplished using a polarised beam splitter (PBS) and a 50:50 beam splitter. After recombination, two beams contain all the necessary frequencies for molecule detection in the $X^2\Sigma^+, (\nu = 0, N = 1)$ state, which then pass through electro-optic modulators (EOMs) before being coupled into fibres and transported to the beamline. The EOMs were used for polarisation modulation during the development of the detection schemes, but they were not present during the laser cooling experiments in chapter 4.

Since the recombination PBS is imperfect, some light is not recombined, which is sent to the wavemeter via fibre for frequency monitoring. Similarly, due to the AOMs' less than 100% efficiency, the zero-order component of the 159 MHz AOM is sufficient for frequency tuning and is directed to a cavity for laser tuning and locking (see section 3.3.4).

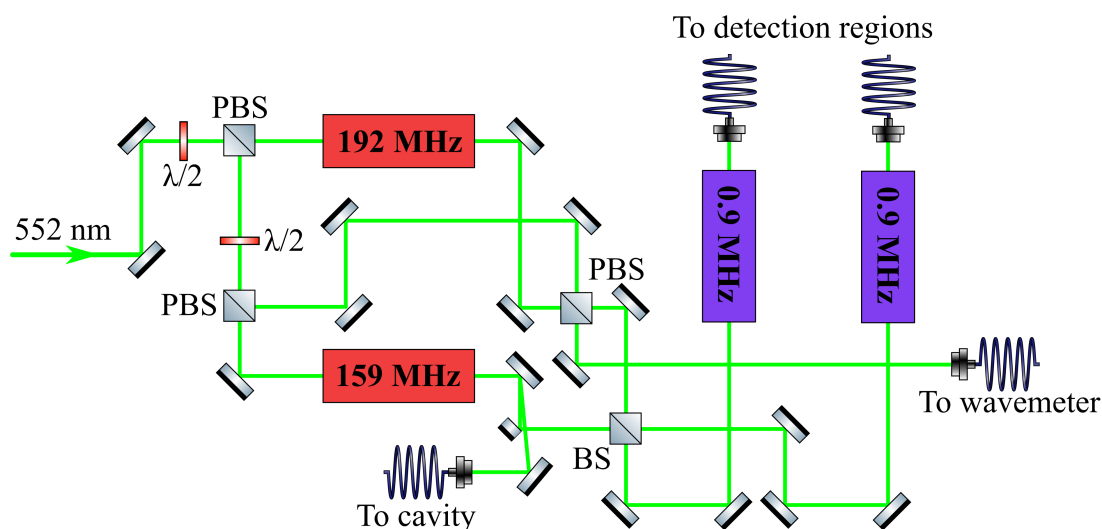


Figure 3.8: Drawing of the probe laser circuit. Labelled components are BS = beam splitter, PBS = polarized beam splitter and $\lambda/2$ = half-wave plate. The red and purple boxes denote AOMs and EOMs, respectively, with their modulation frequencies written inside. At the exits of the optical circuit are several fibres, which take the laser light to various parts of the experiment.

3.3.3 Optical pumping lasers

Chapter 6 focuses on preparing the laser-cooled molecules for the eEDM measurement and explains the new pumping scheme. An energy level diagram illustrating the transitions involved is shown in Figure 6.1. Microwaves are used to couple molecules in the $N = 1$ rotational state to the $N = 2$ rotational state, after which optical laser light is used to pump molecules from the $N = 2$ and $(N = 0, F = 1)$ states to the $(N = 0, F = 0)$ state using the $^{\circ}\text{P}_{12}(2)$ and $\text{Q}(0)$ transitions, respectively. Our experiment uses laser light shared with another YbF eEDM experiment at Imperial College, which has excess laser power. A brief discussion of their laser systems is provided here, and a detailed description can be found in C. Ho's thesis [3].

The laser addressing the $\text{Q}(0)$ transitions is created from a Toptica DL Pro infrared seed laser, which is power amplified using a Keopsys YFA and frequency doubled in free space using a PPLN crystal [3]. Although it outputs around 400 mW of the desired frequency, we share this laser with our colleagues and typically receive around 20 mW of power.

The laser addressing the $^{\circ}\text{P}_{12}(2)$ transitions is generated in a similar manner, with a Toptica DL Pro seed laser being power amplified using a Keopsys YFA and frequency doubled to produce 552 nm laser light. A 185 MHz AOM is used to shift the frequency of some of the light to address the hyperfine structure of the $N = 2$ rotational state. After sharing the laser with our colleagues and transporting it to our lab, we obtain ~ 55 mW of unshifted laser light and ~ 20 mW of 185 MHz-shifted light.

3.3.4 Tuning and frequency locking of laser systems

Frequency locking

To ensure stability, all of our lasers are locked to a Moglabs Cateye ECDL Model CEL 002 reference laser that is shared with other experiments. This reference laser is frequency locked to a 780 nm sub-Doppler feature of rubidium using Zeeman-modulated spectroscopy.

Figure 3.9 shows the optics setup employed in our laser frequency monitoring/locking system, which allows us to compare the relative frequencies of our lasers with a reference laser. The process starts with the frequency doubled light of the cooling laser, probe laser, and first excited vibrational repump laser. These beams are combined with the reference laser light using half-wave plates, polarising beam splitters, and dichroic mirrors. To facilitate the frequency comparison, a Fabry-Perot cavity is used. The combined beams are directed through a Thorlabs SA200-5B visible cavity before being split using the reverse method employed for their combination. Finally, the beams are directed onto amplified photodiodes. By analysing the signals received from the photodiodes, a computer performs a software lock of the frequency, ensuring precise frequency control. It's worth noting that the optical pumping light used in our experiment, borrowed from another YbF eEDM experiment, uses the same model of Fabry-Perot cavity to lock the $Q(0)$ and $^{\circ}P_{12}(2)$ lasers in a similar manner.

The infrared light of the second excited vibrational repump laser is sampled and directed through a Thorlabs SA200-8B infrared cavity, along with some reference light. The process is similar to the previous steps. We utilise a software called "transfer cavity lock" (TCL) to lock our lasers to the reference laser. For more details regarding the software aspects of TCL, please refer to section 3.5.1.

The peak transmission of a cavity depends on the wavelength of the light attempting to traverse it. We obtain a periodically scanned cavity length by varying the cavity length using a sawtooth waveform. The photodiodes each produce a Lorentzian-shaped curve when the light intensity is plotted against the scanning voltage of the cavity. Using Lorentzian curve fitting, TCL precisely determines the voltage for the peak transmission of each laser through the cavity. A software-based proportional-integral (PI) control loop maintains the relative voltage difference between the reference peak and the other lasers by adjusting the grating piezo in each of the seed lasers, thus frequency-locking them to the reference laser.

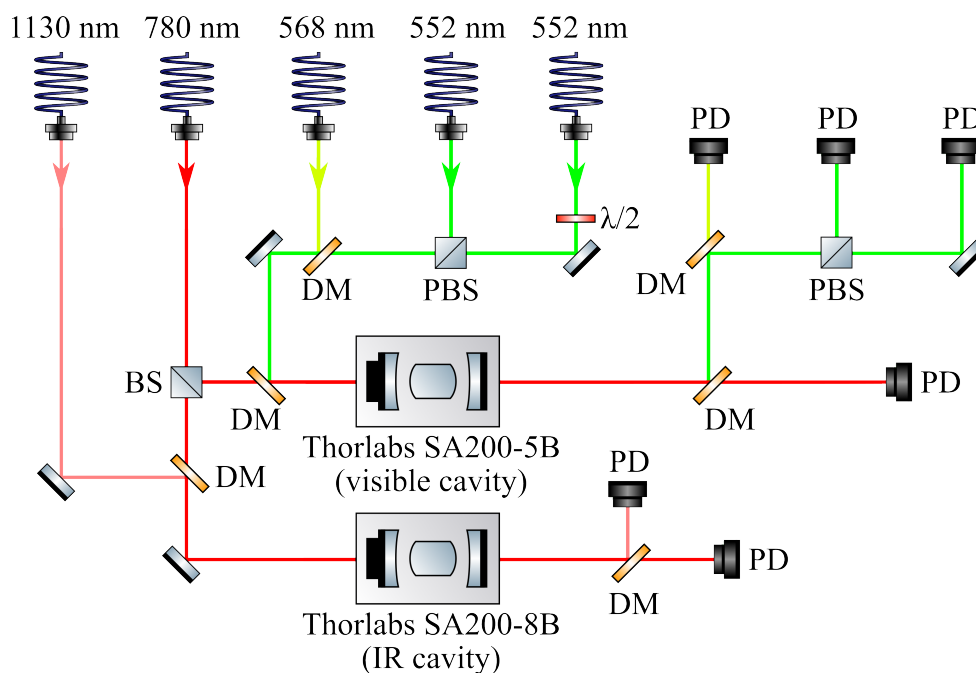


Figure 3.9: Diagram depicting the setup for laser frequency locking. The labelled components include dichroic mirrors (DM), a beam splitter (BS), polarised beam splitters (PBS), a half-wave plate ($\lambda/2$), and photodiodes (PD). The laser light of different frequencies enters the optical circuit through fibres in the figure's top-left corner.

Frequency tuning

We begin tuning laser frequencies by roughly adjusting the current and temperature of the seed laser diodes while monitoring the frequency on a HighFinesse Wavelength Meter WS6-MC-335. We have found that this method is accurate to within ~ 500 MHz, which is less than the free spectral range of the frequency-locking cavities.

Next, we direct the laser light onto the molecular beam and measure the resulting light-induced fluorescence using a PMT. To achieve fine frequency adjustment, we vary the voltage sent to the grating piezo in the seed laser, another TCL functionality. Once we have identified the desired frequency, we lock the laser to the reference laser and then measure the LIF signal as we scan the voltage sent to the grating piezo over an appropriate range. We fit a curve to the resulting peak obtained from the LIF signal. This allows us to determine the precise centre voltage that needs to be applied to the grating piezo, thereby completing the tuning process.

3.4 Microwave systems

Microwaves are used in detecting molecules following an eEDM measurement and transferring molecules from the laser cooling state to the eEDM measurement state. However, as of the writing of this work, only the latter process has been implemented, and thus it will be the focus of our discussion here.

Laser cooling is applied to YbF molecules in the first excited rotational state, $N = 1$. However, for the eEDM measurement, it is necessary to transfer these molecules to the absolute ground state, $X^2\Sigma^+(\nu = 0, N = 0, F = 0)$. This transfer is achieved by coupling the $N = 1$ molecules to the $N = 2$ states using microwaves. Simultaneously, visible laser light is applied to address the $^{\circ}\text{P}_{12}(2)$ and $\text{Q}(0)$ transitions, leading to the pumping of molecules into the $(N = 0, F = 0)$ state. For a more comprehensive understanding of the pumping scheme, please refer to section 6.1, where it is discussed in detail.

Transition(s)	Applied microwave frequency (MHz)	Frequency difference of transitions (MHz)
$(N = 1, F = 0) \rightarrow (N = 2, F = 1)$ and $(N = 1, F = 2) \rightarrow (N = 2, F = 2^+)$	28,966.5	11.0
$(N = 1, F = 1^-) \rightarrow (N = 2, F = 2^-)$ and $(N = 1, F = 1^+) \rightarrow (N = 2, F = 2^+)$	28,935.3	1.8
$(N = 1, F = 2) \rightarrow (N = 2, F = 3)$	28,927.3	-

Table 3.1: Microwave frequencies needed to couple $N = 1$ and $N = 2$. The five transitions can be addressed by three frequencies.

Certain microwave transitions used have frequencies that are close enough to each other that, with sufficient microwave power, the same frequency can drive multiple transitions. Table 3.1 displays the three frequencies necessary to couple all of the $N = 1$ and $N = 2$ hyperfine states. The frequency applied to drive each pair of transitions is adjusted to account for the varying transition couplings. In the right-hand column of the table, you can find the frequency differences between the pairs of transitions.

Figure 3.10 provides a schematic illustrating the generation of microwave frequencies. In this setup, a Windfreak Technologies SynthHD (v2) two-channel microwave synthesiser generates fre-

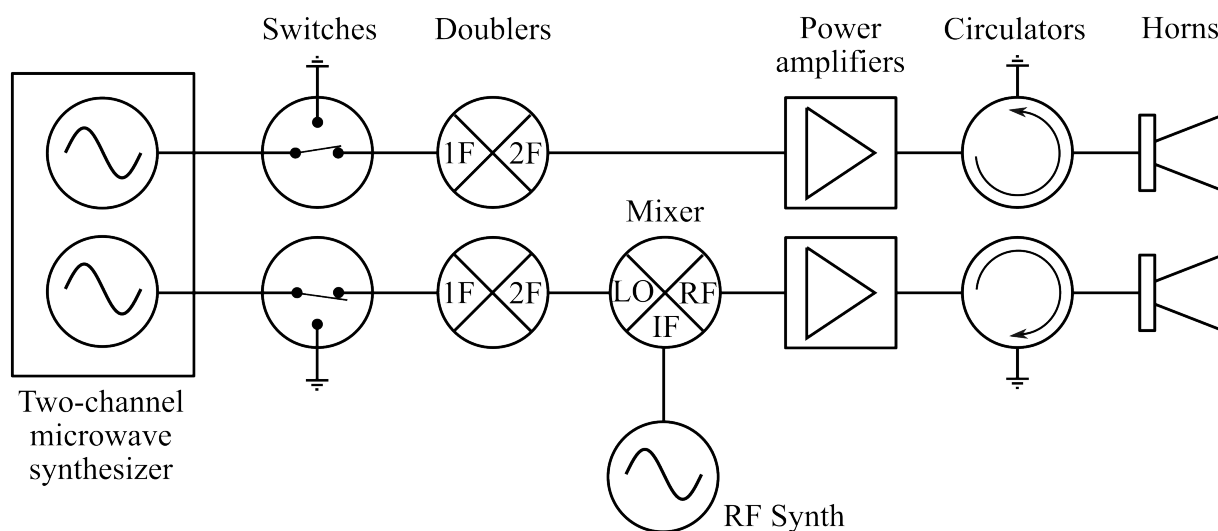


Figure 3.10: Schematic of the microwave setup for optical pumping. Two microwave waveforms are created using a two-channel microwave synthesiser. One of them is frequency doubled (denoted by the 1F and 2F symbols on the inputs and outputs of the doublers) before being power amplified and sent to a microwave horn. The other channel output is frequency-doubled and used as the local oscillator (LO) in a double-balanced mixer. A radio frequency (rf) waveform is applied to the mixer's intermediate frequency (IF) input. The output of the mixer is then power amplified and sent to a microwave horn.

quencies within the range of 10-15 GHz. Each channel can deliver up to 20 dBm of power². Mini-circuits (MSP2T-18-12+) mechanical switches are mounted on the synthesiser outputs to switch the microwaves on and off. They have a switching time of 20 ms, which is too slow for pulsing the microwaves during a single pulse of molecules but useful for switching in between molecule bunches.

In figure 3.10, the top output of the microwave synthesiser is used to address the $(N = 1, F = 2) \rightarrow (N = 2, F = 3)$ transition, outputting a frequency of 14.4636 GHz. This is frequency doubled using a Marki Microwave MMD-1030H double-balanced doubler. At the desired frequency, the doubler outputs a 28.9273 GHz waveform of power³ up to ~2 dBm. The output power of the synthesiser limits the output power of the doubler. After doubling, the waveform is amplified by an Erzia ERZ-HPA-2400-3000-37 high power amplifier⁴ (HPA). The HPA can amplify a waveform up to 50 dB,

²The maximum power output of the microwave synthesiser depends on the specific frequency of the generated waveform. For instance, a waveform at 14.5 GHz was measured to have a maximum power output of approximately 14 dBm.

³Power measurements of some waveforms were extrapolated from other measurements because our spectrum analyser is limited to signals up to 26 GHz.

⁴At the time of writing this, only one of the two high power amplifiers is operational. This affected the measurements which could be made and are discussed in detail in chapter 6.

with a maximum output power of around 35 dBm for a 29 GHz input waveform. The waveform then propagates through a Pasternack PE83CR1028 circulator to a Pasternack PE9850/SF-20 microwave horn mounted inside the beamline (i.e. in vacuum). The circulator redirects back reflections to a microwave terminator, protecting the HPA output stage from potential damage.

The other synthesiser output channel in figure 3.10 is used to address the two pairs of transitions from table 3.1. It outputs a 14.47535 GHz waveform which is frequency doubled in the same manner as the other channel. This is then used as the local oscillator for a Marki Microwave MM1-0832H double-balanced mixer. The intermediate frequency of the mixer is a 15.6 MHz radio frequency waveform, provided by an Analog Devices AD9850 direct digital synthesiser⁵ (DDS). Two sidebands are created with a frequency difference of ± 15.6 MHz relative to the LO frequency (which is highly isolated from the RF output of the mixer). The output of the mixer is handled in an identical manner as the other channel is: power amplifier, circulator and microwave horn. With the HPA supplying energy to two waveform frequencies, it provides less amplification. Typically, each of the two frequencies leave the HPA with around 27 dBm of power. The microwave horn for this waveform is mounted in the vacuum chamber with its antenna orthogonal to the other horn.

3.5 Computer control

A computer predominantly controls the experimental apparatus. The functionalities encompass a range of tasks, including but not limited to: tuning and locking lasers using TCL; control of the molecular source; controlling switches, shutters and other elements in different patterns for various experiments. In this section, I describe these core pieces of software. It is worth noting that all the relevant software discussed here can be accessed in the CCM GitHub repository [102].

⁵The DDS integrated circuit device is mounted on a breakout board and controlled using a homebuilt microcontroller circuit.

3.5.1 Transfer cavity lock

Transfer Cavity Lock (TCL) is our custom software designed to control the many laser systems in our experiment. It is used for the tuning and frequency locking of our lasers, as discussed in section 3.3.4. TCL interacts with the experiment through a dedicated National Instruments multifunction input/output module, receiving inputs such as the cavity voltage ramp trace and the amplified photodiode outputs. Through user-defined inputs, the software allows precise control over the parameters of the frequency lock. Notably, TCL governs the voltage sent to the grating piezos of the various lasers in our setup, enabling precise adjustment of their frequencies. One limitation of TCL, however, is its refresh rate for the frequency lock, which currently operates at a few hertz. While this has not posed any issues thus far, it could potentially become a limitation when attempting high-precision measurements of the eEDM.

Figure 3.11 displays a screenshot of the TCL interface. The top portion of the interface shows the cavity voltage ramp waveform, while the bottom portion presents the lock error signal. In the middle section, multiple photodiode signals can be selected for display. The screenshot shows the reference laser signal and the probe laser signal. If the user checks the "Lock" checkbox, the software will attempt a lock between the laser signal and its corresponding reference laser signal. Even when switching to another tab, the laser remains locked. Additionally, the interface provides controls for adjusting the voltage sent to the grating piezos in the lasers, enabling precise frequency control of the lasers.

3.5.2 Scan master

Control of the measurement processes is achieved using a custom software tool called Scan Master (SM). It uses a National Instruments pattern generator and data acquisition modules, allowing us to orchestrate experimental procedures and capture measurements with microsecond precision. With SM, we can define user inputs to control the timing of various events, such as molecule creation, triggering optical beam shutters, acquiring signals from PMTs, and synchronising camera triggers.

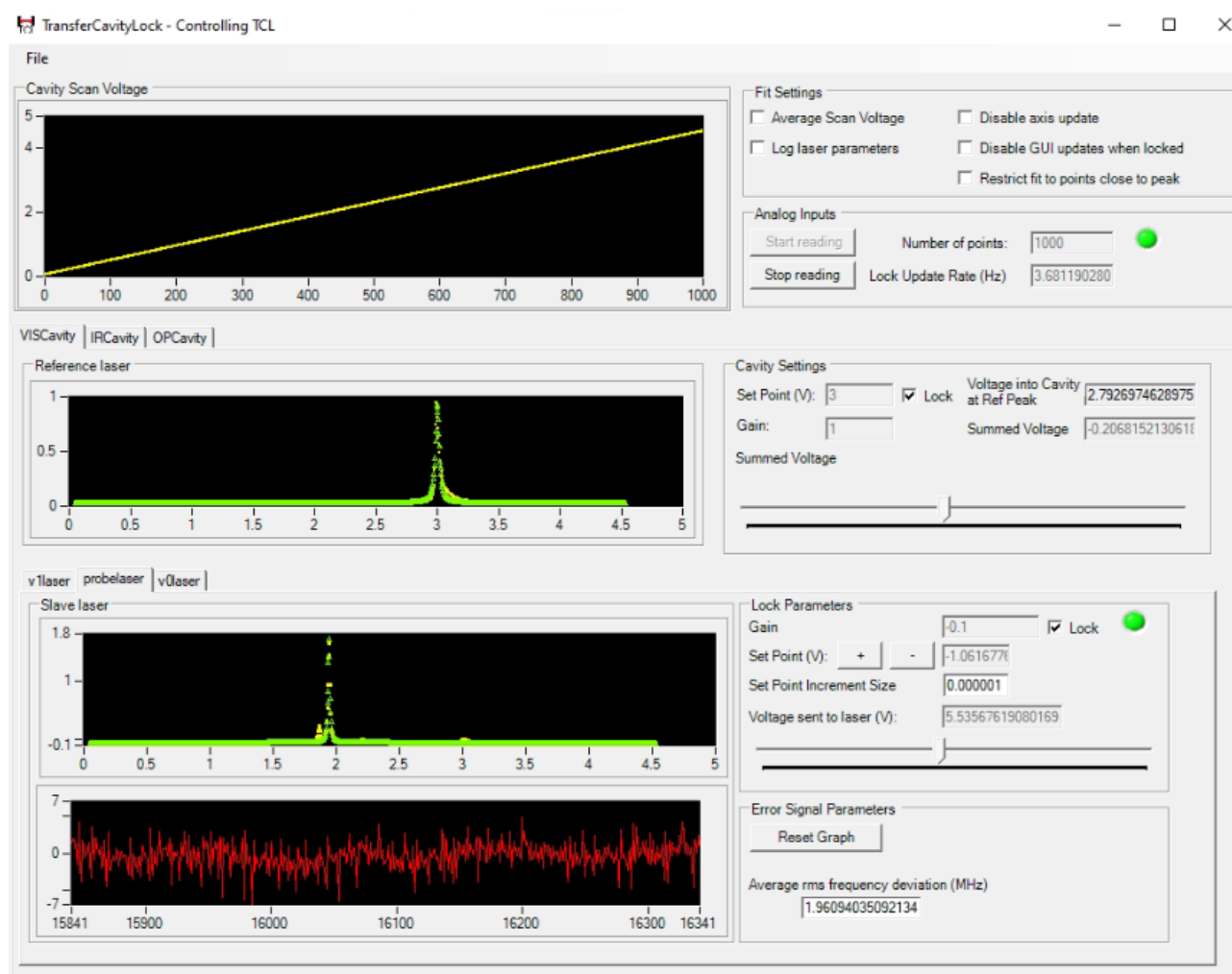


Figure 3.11: Screenshot of the TCL interface. The top panel shows the ramp waveform sent to the cavity, depicting the voltage applied over time. At the bottom, a plot of the lock error signal is displayed, indicating the laser lock status. The remaining panels showcase the photodiode signals for different lasers, illustrating their response as a function of the applied voltage to their respective cavity. The overlaid green points represent Lorentzian fits to the yellow data.

One of the key features of SM is its ability to facilitate quick parameter adjustments and parameter scanning. For instance, in the context of fine-tuning laser frequencies (as discussed in section 3.3.4), SM interfaces with TCL to perform precise scans of frequency locks while simultaneously measuring light-induced fluorescence from the molecules using a PMT. This versatility in scanning parameters across different measurements is what inspired the software's name, Scan Master.

3.5.3 Hardware controller

The control of lasers and measurements is primarily accomplished through the use of TCL and SM, as discussed in sections 3.5.1 and 3.5.2, respectively. Additionally, a software component known as the "hardware controller" is responsible for managing various other components within the experimental setup⁶. While most hardware interactions are facilitated through a dedicated National Instruments multifunction input/output module, several USB and RS232 connections are also employed to communicate with specific devices.

The hardware controller plays a crucial role in automating the control of the molecular source, a frequently employed function. It carries out temperature measurements of various source components and pressure measurements of both the source and beamline. This information serves as input for the automated process of refreshing the cryocooled source, as detailed in section 3.2.2. The user can define specific parameters to control the timing of this process while the graphical interface displays temperature and pressure measurements over time.

The refresh process initiates by heating the cryocooler to desorb the buffer gas from the charcoal sorption pumps. Uncontrolled heating can cause the vacuum pressure in the source chamber to rapidly increase by several orders of magnitude within seconds. Such elevated pressures can result in thermal coupling between the cold components and the room temperature chamber, leading to an uncontrolled desorption/sublimation of gases. To prevent this, the vacuum pressure is continuously monitored at intervals of 100 ms, and the heaters are promptly turned off if the pressure exceeds a predefined limit, typically around 1×10^{-4} mbar. During this period, the turbomolecular pumps are given time to reduce the pressure. The heaters are reactivated once the pressure falls below the limit, and the cycle repeats until all the buffer gas has desorbed. This iterative process continues until the charcoal surfaces reach a user-defined temperature, typically 40 K.

Subsequently, the cryocooler is deactivated, and the heaters warm the source to a user-defined target temperature, typically a few degrees above room temperature. While pressure limits remain in place during this stage, they are rarely exceeded⁷. Only a small quantity of residual background

⁶In the CCM GitHub repository, the hardware controller is referred to as "UEDMHardwareControl" [102].

⁷It is important to closely monitor the pressure in the vacuum chamber until it reaches room temperature to detect

gases should remain to sublime from the cold surfaces. After a predetermined duration, the heaters are deactivated, and the cryocooler is reactivated. Importantly, this process's heating and cooling phases can also be independently performed. This capability proves useful, for instance, when opening the source for maintenance the following day or concluding maintenance and intending to cool down the source overnight.

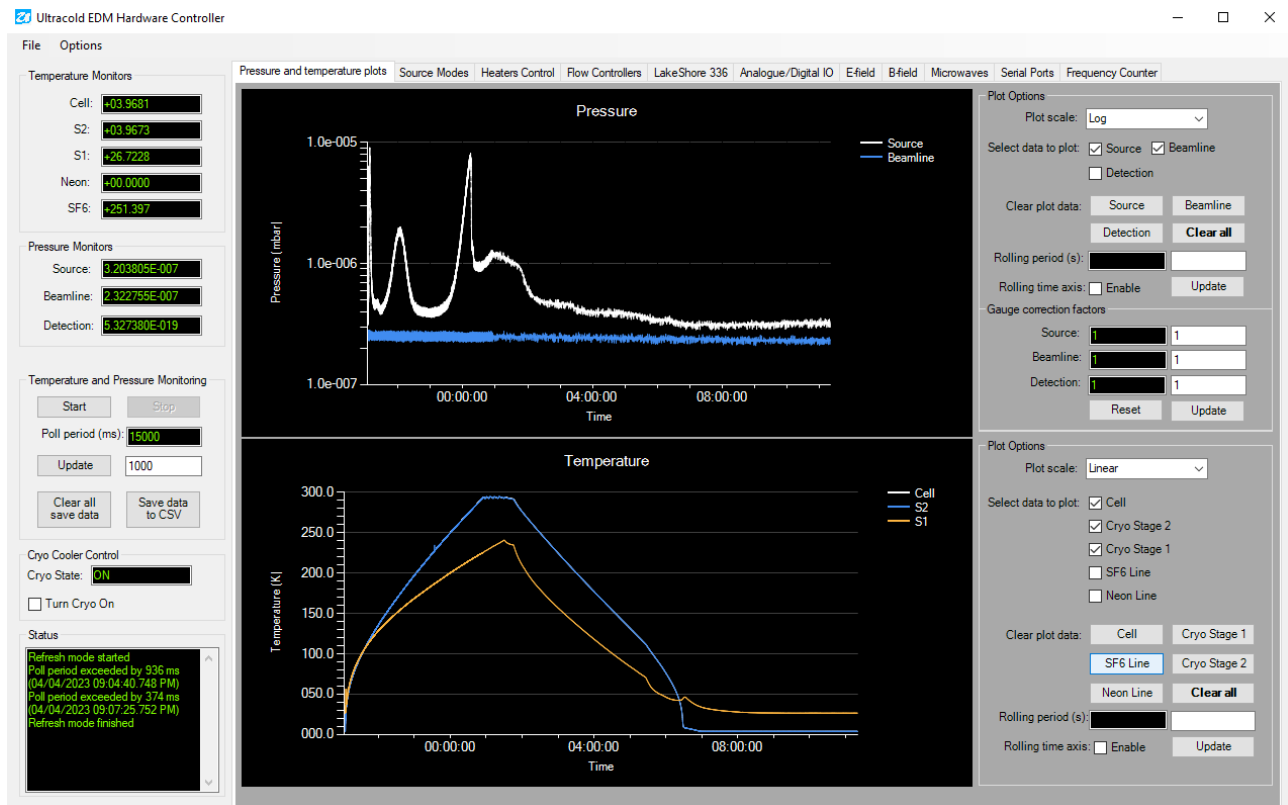


Figure 3.12: Screenshot of the hardware controller interface. The left-hand panel displays the most recent temperature and pressure measurements and allows the polling period to be adjusted. The main panel features multiple tabs for controlling different experiment components. The currently selected tab shows pressure and temperature measurements plotted over time. The presented data showcases an overnight source refresh process.

Figure 3.12 presents a screenshot of the hardware controller interface. The controller's functionalities are organised into multiple tabs at the top of the screen, while the left-hand panel, which displays the latest temperature and pressure measurements, is always visible. The polling period can also be adjusted from the left-hand panel. The selected tab shows the temperature and pressure measurements over time, showcasing data from an overnight source refresh. The bottom plot shows the temperatures of various source components, demonstrating their gradual warming up and subsequent cooling down throughout the night. The top plot depicts the pressure in the any vacuum leaks or frozen air on cold surfaces.

source and beamline during the overnight process. The white curve represents the source pressure, which rises due to the desorption and sublimation of gases when specific temperatures are reached. In contrast, the blue curve representing the beamline pressure remains unchanged as the gate valve connecting the two vacuum chambers is closed during this procedure.

The hardware controller not only manages the refreshing process of the molecular source but also controls several other essential components of our experiment. These include gas flow controllers, microwave and radio frequency synthesisers, temperature controller PID loops, and the electric field plates used in the interaction region. The hardware controller plays a critical role in automating and coordinating these various elements, allowing us to dedicate our focus to the physics of the experiment rather than spending time on manual equipment control.

Chapter 4

Laser cooling of the YbF molecular beam

Laser cooling of YbF molecules is a critical component of our improved eEDM measurement scheme. In 2018, researchers at Imperial College London demonstrated the first one-dimensional transverse laser cooling of a YbF beam [80], paving the way for the development of our current two-dimensional transverse laser cooling apparatus [1]. This chapter presents the results of experiments on one-dimensional and two-dimensional transverse laser cooling of YbF molecules. Firstly, the laser cooling scheme, apparatus design, and measurement sequence are outlined in detail, building on the more general discussions presented in chapters 2 and 3. Experimental results are then provided for both one-dimensional and two-dimensional laser cooling of YbF. Finally, the two-dimensional laser cooling results are used to calculate the brightness of the molecular beam and to estimate the statistical uncertainty associated with an eEDM measurement made using this improved molecular beam.

4.1 Laser cooling of YbF

To describe the laser cooling scheme, this chapter relies upon the theoretical foundations established in chapter 2. Specifically, the energy level structure for YbF described in section 2.1 and the principles of laser cooling of atoms and molecules outlined in section 2.3 serve as the basis for the detailed description of the laser cooling scheme provided in this section.

The energy level diagram in figure 4.1 illustrates the YbF laser cooling scheme, highlighting the ground state $X^2\Sigma^+$ and the first excited state $A^2\Pi_{1/2}$. The diagram shows the first four vibrational states of the ground electronic state and only the $v' = 0$ and [561] states of the excited electronic state. The solid arrows represent the addressed optical transitions, with the hyperfine structure of the $v = 0$ state separated for clarity. The branching ratios of the excited states are indicated by wavy arrows pointing towards the target decay state, with accompanying probabilities of decay indicated next to each arrow.

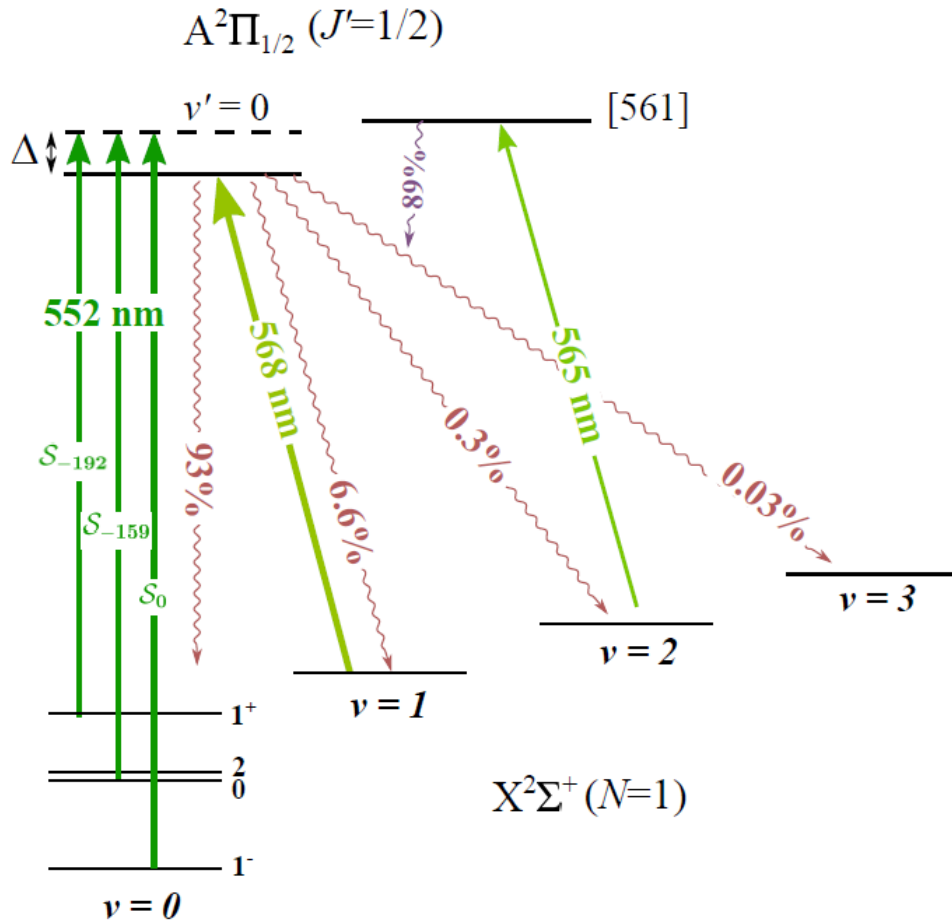


Figure 4.1: Energy level diagram illustrating the laser cooling scheme for YbF. The electronic ground state and first excited state are labelled as $X^2\Sigma^+$ and $A^2\Pi_{1/2}(J' = 1/2)$ respectively. The first four vibrational states are shown in the ground electronic state, while only the $v' = 0$ and [561] states are shown for the excited electronic state. Solid arrows depict the optical transitions, while wavy arrows indicate the relevant branching ratios. The hyperfine structure of the ground vibrational state is also shown, with optical components labelled S_δ , where δ is the detuning (in MHz) relative to the highest frequency sideband. The cooling frequencies are detuned by Δ from the excited state, $A^2\Pi_{1/2}(J' = 1/2)$, $v' = 0$. The hyperfine structure is not shown for the higher vibrational states. Figure reproduced from Alauze et al. (2021) [1].

Cooling takes place via the transition $|X^2\Sigma^+, \nu = 0, N = 1\rangle \rightarrow |A^2\Pi_{1/2}, \nu' = 0, J' = 1/2\rangle$, which has a natural linewidth of $\Gamma = 2\pi \times 5.7$ MHz. This excited state has a lifetime of 28(2) ns, facilitating the scattering of many photons in a short time [64]. The ground and excited states comprise four and two hyperfine components, respectively. In the ground state, the $F = 0$ and $F = 2$ hyperfine components are only separated by 6 MHz whilst the excited state hyperfine components have a splitting of only 3 MHz. As a result, only three optical frequencies are required to address the cooling transition. Relative to the highest frequency transition, the two other frequencies are detuned by -159 MHz and -192 MHz - hence their labels S_0 , S_{-159} and S_{-192} . The three frequency components are not necessarily equally weighted in intensity, as discussed in section 4.3.1. To achieve sub-Doppler cooling, all three frequencies are blue-detuned¹ by the frequency Δ and used with magnetic fields to induce the two sub-Doppler laser cooling mechanisms described in section 2.3.

The cooling transition is rotationally closed, as electric dipole transitions from the cooling excited state to higher rotational levels of the ground electronic state are forbidden. However, this is not the case for decay to higher vibrational states, for which the branching ratios are non-negligible up to $\nu = 3$. Although the branching ratio for decay back to the ground state of the cooling transition is 93%, after only a few cycles, many molecules will be lost to higher vibrational states. To mitigate this effect, molecules must be repumped back into the cooling cycle from higher vibrational states. Using extra lasers, molecules can be repumped from these states back into the cooling cycle. Empirically, we have found that the $\nu = 3$ repump does not make a significant difference. A 568 nm laser (with sidebands added to address hyperfine structure) is used to repump molecules from the $\nu = 1$ state directly to the cooling excited state. Molecules are repumped from the $\nu = 2$ state via the [561] state, which predominantly decays to the $\nu = 1$ state.

4.2 Experimental setup

This section provides an overview of the experimental setup employed in the YbF laser cooling experiments. Firstly, the experiment is introduced. Next, the laser cooling region is described in

¹When blue-detuned, the laser light is of greater frequency than the resonance frequency of the transition. Conversely, red-detuned light would have a frequency less than resonance.

detail, including the optical molasses optics and magnetic fields. Following this, the clean-up region is briefly discussed before a detailed discussion of molecule detection and the measurement sequence.

4.2.1 Overview of the YbF laser cooling experiment

In section 3.1, an overview of the YbF eEDM measurement apparatus is provided. To optimize the laser cooling setup and characterize the beam, a shorter beamline was utilised for the laser cooling experiments. Figure 4.2 depicts the beamline configuration during the laser cooling experiments. The laser cooling experiment began by producing a YbF molecular beam directed down the z-axis beamline. The molecules then travelled through the laser cooling region and a clean-up region, where any molecules remaining in excited vibrational states were repumped back to the cooling cycle. Finally, the molecules were detected by two laser-induced fluorescence (LIF) detectors, positioned at different z-axis locations. The first detector viewed the molecular beam from the side of the beamline, while the second detector viewed the molecular beam end-on, as the molecules moved towards the camera.

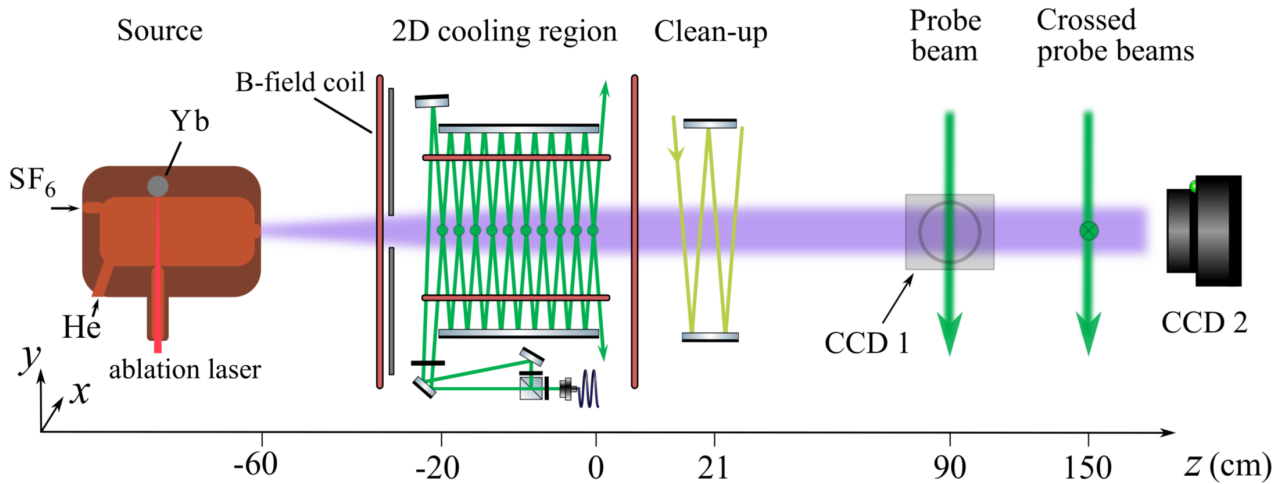


Figure 4.2: Diagram of the YbF laser cooling experiment. Molecules were created using a buffer gas source and propagated down the beamline in the z-axis. They traveled through cooling light in the presence of a magnetic field provided by Helmholtz coils. After laser cooling, clean-up light repumped molecules that remained in the $\nu = 1$ and $\nu = 2$ vibrational states. Finally, the molecules passed through spatially separated LIF detectors. Figure reproduced from Alauze et al. (2021) [1].

4.2.2 Laser cooling region

Figure 4.3 shows the "panoramic"² vacuum chamber that was used for the laser cooling experiments. The two transverse axes were referred to as the horizontal and vertical cooling axes. For each axis, cooling light was delivered to the beamline via optical fibre and was immediately split on a polarising beam splitter (PBS) before being directed into the vacuum chamber, as shown in figure 4.2. The beams reflected back and forth along 20 cm long mirrors, making a total of 15 round trips. At the crossing points of the beams, intensity-varying or polarisation-varying standing waves were formed depending on the relative polarisation of the two beams. A photo of the horizontal cooling beams is shown in figure 4.3. The two beams formed sets of interleaved spots where the light scattered on the panoramic chamber window and mirror. Care had to be taken when installing the windows to avoid stress-induced birefringence, which could result in unintended rotation of the cooling light polarisation³.

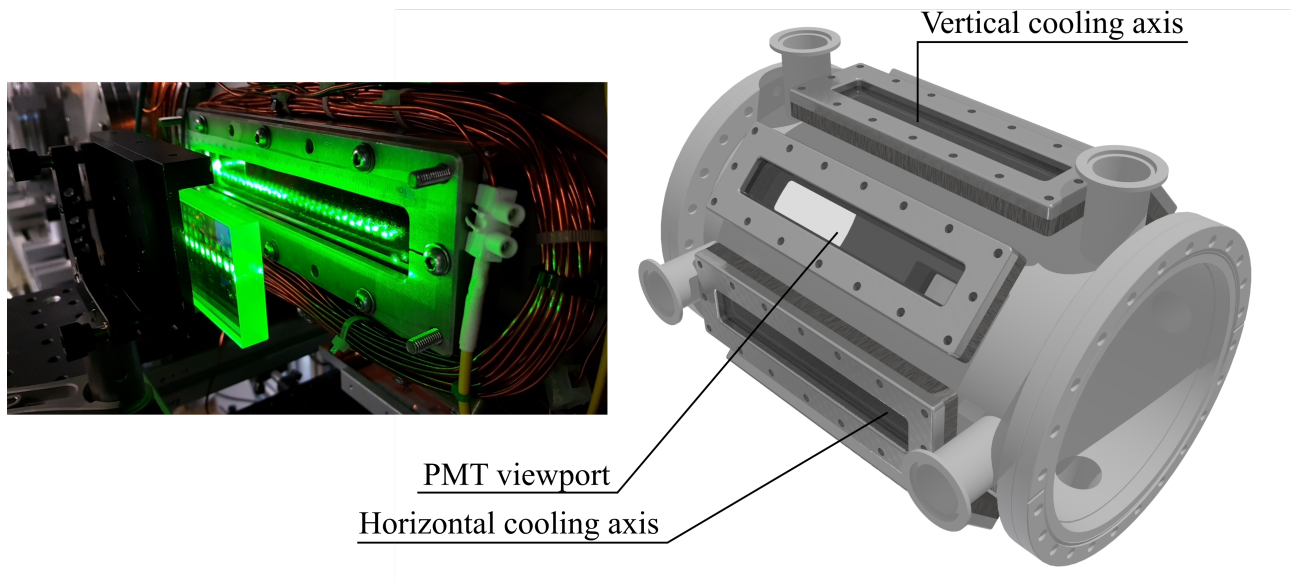


Figure 4.3: Panoramic vacuum chamber for two-dimensional laser cooling. The photo shows cooling light on the horizontal axis. Enamelled copper wire is wrapped around the window flange; it is one of two magnetic field coils for the x-axis, with its counterpart wrapped around the opposing flange.

Control of magnetic fields was achieved using three pairs of electromagnets. Two pairs were formed

²The name "panoramic" refers to the wide range of optical access provided by the vacuum chamber.

³Unintended rotation of cooling light polarisation can be observed through the lack of sensitivity of the sub-Doppler cooling mechanisms to the applied magnetic field strength.

by winding enamelled copper wire around each of the four cooling light window flanges, which provided magnetic fields in the x and y axes. One of the coils can be seen in the photo in figure 4.3. Due to their wide separation, these pairs of coils were not arranged in the Helmholtz configuration, resulting in some non-uniformity near the start and end of the cooling region and away from the beamline axis. In the z-axis, a pair of 1 m square coils were arranged in the Helmholtz configuration and centred on the cooling region. The three pairs of coils were used to cancel the background field in the cooling region and apply a uniform field.

4.2.3 Detection

Two cameras, labelled CCD1 and CCD2, were used to detect molecules. Both relied on light-induced fluorescence for detection but were positioned differently relative to the molecular beam. Both cameras were fitted with spectral bandpass filters, which allowed the 552 nm detection light to be transmitted but blocked any vibrational repump light that might scatter down the beamline. Only CCD2 was needed to measure the number of molecules which had been laser-cooled, while two detectors were required to measure the molecules' transverse temperature. Details on this temperature measurement method are discussed in the two-dimensional laser cooling results section 4.4.4.

CCD1, a Hamamatsu EMCCD C9100-23B, was positioned 90 cm from the end of the laser cooling region and viewed the molecular beam from the side, giving it a field of view in the y-z plane. As a result, CCD1 could not measure the molecular beam's x-axis spatial distribution. The probe light was directed downwards, travelling in the negative y-direction, allowing only the y-axis spatial distribution to be measured. Figure 4.4 displays two example images from CCD1. Both images were created by averaging 507 photos and subtracting the averaged background. The laser-cooled molecules are clearly visible in the centre of the left-hand image, while the uncooled molecular beam is seen as approximately uniform in the y-axis of the probe beam. Typically, the images were cropped to the regions outlined by red rectangles, and the signal was integrated along the z-axis.

The second camera, CCD2, was an Andor iXon 888. It was placed 60 cm from CCD1, at a distance

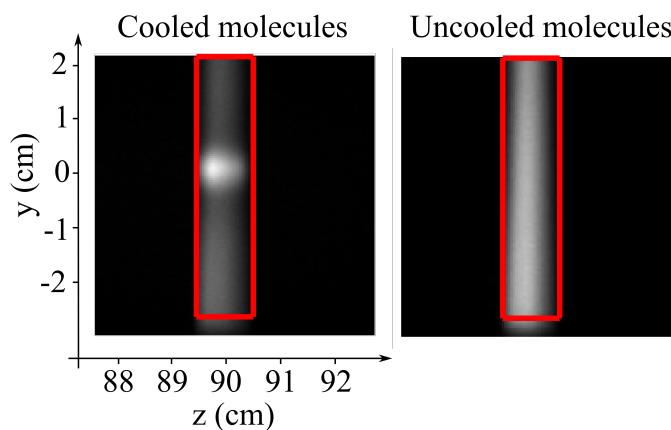


Figure 4.4: Example images from CCD1. Both are averages of 507 photos with the averaged background subtracted. On the left, laser cooling has been applied to the molecular beam; the bright spot in the centre is the cooled molecules. The right-hand image is of the uncooled molecular beam. Red rectangles denote the region of interest for our analysis.

of 150 cm from the end of the laser cooling region. To allow measurement of the molecular beam's spatial distribution in both the x and y axes, the camera was positioned at the end of the beamline, with the molecules moving towards it and a field of view in the x - y plane. The probe light entered the chamber in the positive x -direction and was looped around to pass through the chamber again in the negative y -direction, forming a cross-shaped probe beam. Figure 4.5 displays two example images from CCD2, which are both averages of 507 photos with the averaged background subtracted. The cross-probe beam configuration is easily discernible where molecules are present. In the left-hand image, the ultracold molecules appear as a bright spot at the center of the cross. The blue and red rectangles denote the regions of interest for horizontal and vertical laser cooling, respectively, and their intersection is used for two-dimensional laser cooling analysis.

The cross-probe beam configuration was advantageous when aligning the cooling beams. Consider the following example. If one-dimensional laser cooling were applied on the horizontal axis, one would expect the ultracold molecules to be widely distributed along the y -axis, with a narrow x -axis distribution. In figure 4.5, this would appear as a bright stripe inside the blue rectangle. However, if the cooling beams were misaligned, the cooled molecules would be off-centre on the x -axis - potentially outside the blue rectangle. If a probe beam was only propagating in the y -axis, they could miss it entirely, leaving no diagnostic information to correct the problem. The same is true if the x and y axes are reversed in this example. By having a probe beam in both axes, fine-

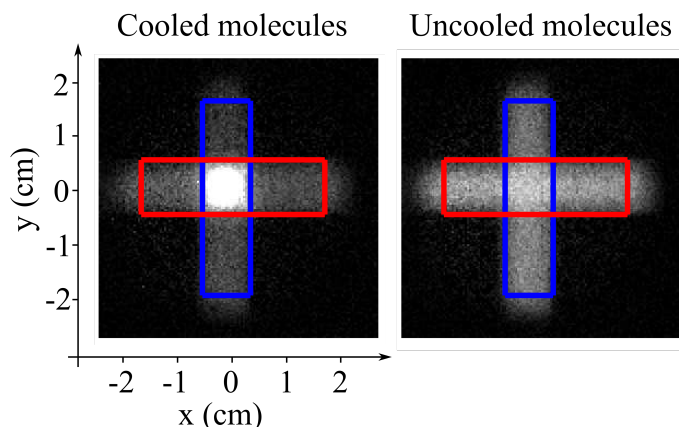


Figure 4.5: Example images from CCD2. Both are averages of 507 photos with the averaged background subtracted. On the left, laser cooling has been applied to the molecular beam; the bright spot in the centre is the cooled molecules. The right-hand image is of the uncooled molecular beam. Red and blue rectangles denote the region of interest for our analysis.

tuning of the cooling beams' alignment is facilitated. In the camera images, this appears to move the ultracold molecules to the centre of the beamline.

4.2.4 Measurement sequence

In general, it is challenging to scatter enough photons from molecules to be able to photograph them. Despite using highly sensitive cameras and minimising background light sources, obtaining high-quality images still required the use of averaging and background subtraction techniques. This section provides an overview of the steps involved in a typical laser cooling measurement.

The process of a laser cooling measurement involved capturing multiple photos of the molecular beam. Below is an example of the set of four photos that were typically taken:

1. Molecules are created and laser-cooled
2. No molecules are created (background photo)
3. Molecules are created, but no cooling light is applied
4. No molecules are created (background photo)

Each photo set consisted of photos of laser-cooled and uncooled molecules (photos 1 and 3), with a corresponding background photo for each (photos 2 and 4) to allow for background subtraction. The laser cooling light was turned off for all photos before the camera was triggered. This set of four photos formed the basis for all laser cooling measurements. To achieve the desired image quality, multiple photo sets were taken and averaged, with 250-500 photo sets per measurement depending on the daily variation in the number of molecules produced by the source.

To operate the other camera, the same process is followed. However, the cameras cannot detect the same molecules because detection occurs on the same molecular transition, and many of the molecules would be pumped into states dark to the pump light by the first detector. Therefore, the two cameras take turns measuring the molecular beam by taking photo sets one after the other.

4.3 Experimental results: one-dimensional laser cooling

Laser cooling of the molecular beam was first optimised separately for each transverse direction. The optimal cooling light detuning was particularly interesting since laser cooling of YbF molecules had never been performed with such high-intensity cooling light. As the cooling light detuning varies, the detuning frequency uniformly shifts all sidebands. Therefore, an investigation into the optimal power balance of the sidebands was performed. This is discussed in section 4.3.1. A measurement of 1D laser cooling with varying detuning is presented in section 4.3.2, where the optimal sideband power configuration was used. All measurements in this section were made with only CCD2.

Measurements were made for each laser cooling configuration, as described in section 4.2.4. This resulted in two images: one of the laser-cooled molecules and one of the uncooled molecules. We averaged the photos and used the standard error of the mean as the uncertainty in the pixel counts. Figure 4.6a presents an example 1D laser cooling data set. The pixel counts were integrated along the y-axis and plotted against the x-axis position. In blue are the uncooled data, whilst in black are the cooled data. The Gaussian-shaped peaks differ in amplitude and width - the cooled peak is taller and narrower. Figure 4.6b shows the ratio of the two data sets with a Gaussian fit

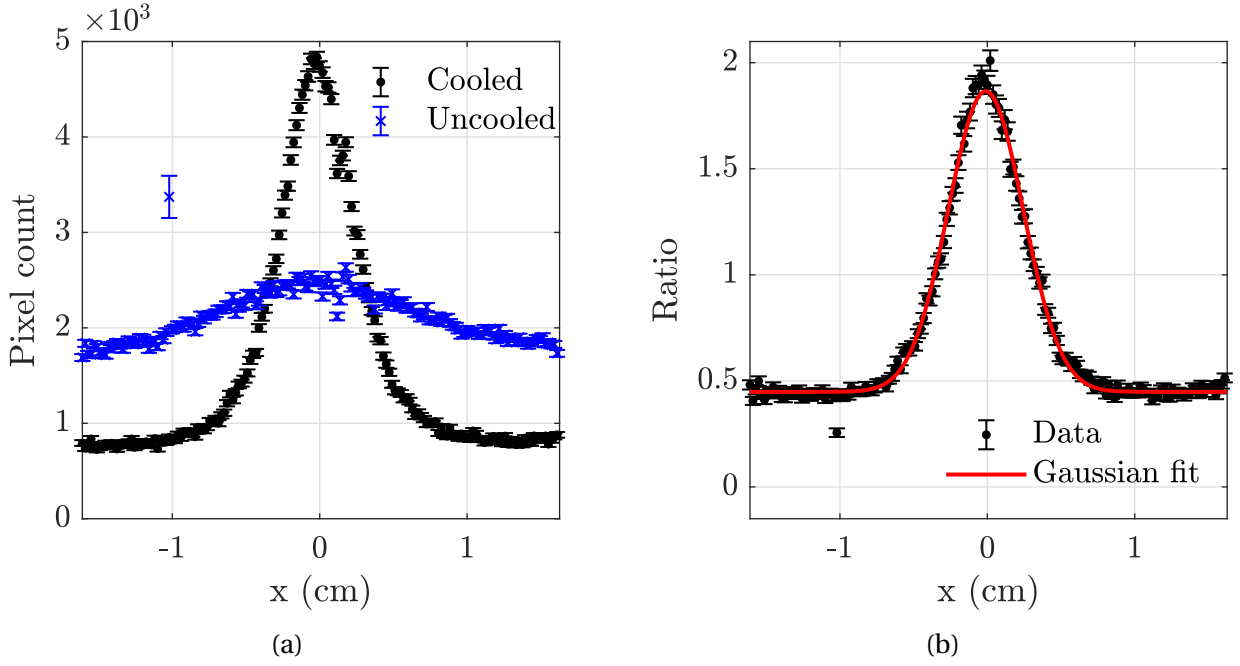


Figure 4.6: Example CCD2 data set with and without 1D laser cooling applied. The left plot (a) displays the pixel counts integrated along the y-axis as a function of position in the x-axis, with black dots representing the cooled molecular signal and blue crosses representing the uncooled molecular signal. The standard error of the mean was used as the uncertainty in the pixel counts. The right plot (b) displays the ratio of the cooled and uncooled molecular integrated pixel counts as a function of position in the x-axis, with black dots representing the data and a red line representing a Gaussian fit with adjusted R-squared = 0.99.

$g(x) = \frac{A}{2\pi} e^{-(x-x_0)^2/(2\sigma^2)} + b$ overlaid. The area of the peak in our results is a direct indicator of the number of ultracold molecules that we were able to generate through laser cooling. Two key factors determine the width of the peak. Firstly, the laser-cooled beam's temperature determines the beam's divergence, subsequently affecting the width of the peak. Secondly, only molecules with a velocity below the critical velocity can be effectively cooled, as discussed in section 2.3.4. As a result, the size of the laser-cooled beam that exits the laser-cooling region is determined by the critical velocity. Finally, the offset of the peak from zero is measured by the ratio's baseline, b .

To optimise the laser cooling setup, a figure of merit was required. The number of detected molecules was proportional to the pixel counts, and the area of the fitted Gaussian approximated the number of ultracold molecules. After removing the offset b , the integral of $g(x)$ became $A\sigma$, which was made dimensionless by dividing by a length L . We used the resulting quantity $S_{1D} = A\sigma/L$ as a relative measure of the number of ultracold molecules for different laser cooling experiments. Although the choice of L was arbitrary, we selected $L = w$, where w was the width of the Gaus-

sian probe beam with an intensity distribution of $I(r) = I(0)e^{-2r^2/w^2}$. When $S_{1D} = 1$, the number of ultracold molecules was equivalent to the case without laser cooling. A value greater than one indicated that more ultracold molecules were present compared to the absence of laser cooling.

We expected the baseline, b , to be close to one, as it represents the ratio of the number of cooled to uncooled molecules in the wings of the distribution where no laser cooling occurs. However, our measurements show b in the range of 0.4-0.7 for various experimental conditions. Measurements where $b < 1$ mean fewer molecules are detected when cooling is applied than when not; these molecules are dark to the detection light. Given that we apply sufficient laser intensity to completely repump cooled molecules into the detection state, we surmise that they must have left the cooling cycle. The leading hypothesis for the cause of the leak out of the cycle is described in brief here; for an in-depth discussion, refer to Zhang et al. (2022) [103].

Several states lie between the ground and excited electronic states used for cooling. These low-lying electronic states arise from inner-shell excitation of a 4f electron centred on the Yb atom. Because this results in a vacancy in the 4f shell, they are denoted as $4f_{J,\Omega}^{-1}$. There are two sets of states, with total electronic angular momentum $J = 5/2, 7/2$, separated by an enormous spin-orbit interaction energy [103]. The set with $J = 7/2$ lie below the $X^2\Sigma$ and the $A^2\Pi_{1/2}$ states, and so are the ones of most concern. There are 4 of these states, distinguished by Ω , which is the projection of J onto the internuclear axis. In particular, the decay from the $A^2\Pi_{1/2}$ state to the $4f_{7/2,1/2}^{-1}$ state has a calculated branching ratio of 0.05%. This is at least twice the branching ratio to the $\nu = 3$ ground vibrational state and limits the average number of photons scattered before molecules exit the cooling cycle to around 2000 [103].

Whilst this number of scattered photons is sufficient for laser cooling our molecular beam, it does reduce the time-averaged molecular flux and therefore decreases the sensitivity of an eEDM measurement. A promising proposal is to optically pump the lost molecules to the mixed states [557] or [561] using two lasers with wavelengths close to 1 μm , potentially increasing the number of photons that can be scattered by a factor of 5 [103].

4.3.1 Hyperfine structure of the cooling state

We focus exclusively on sub-Doppler cooling, which uses blue-detuned light as explained in section 2.3.4. The range of detunings that can be used is constrained by the hyperfine structure. Figure 4.7 shows three energy level diagrams of the ground state of the cooling transition. The dashed lines depict the cooling light detuning gradually increasing from left to right. On the left, the cooling light is near to resonance - only a small blue detuning. In the middle diagram, the detuning has increased. The frequency of the S_{-192} sideband sits between the $F = 1^+$ and $F = 2,0$ energy levels, and whilst blue-detuned by $\Delta = 3\Gamma$ from the $F = 1^+$ component, it is also red detuned by 3Γ from the $F = 2,0$ components. As the overall detuning increases to $\Delta = 6\Gamma \approx 34$ MHz, the S_{-192} sideband frequency becomes resonant with the transitions from $F = 2,0$. Since the sub-Doppler cooling mechanisms rely on the light being blue-detuned from resonance, this scenario is detrimental to cooling.

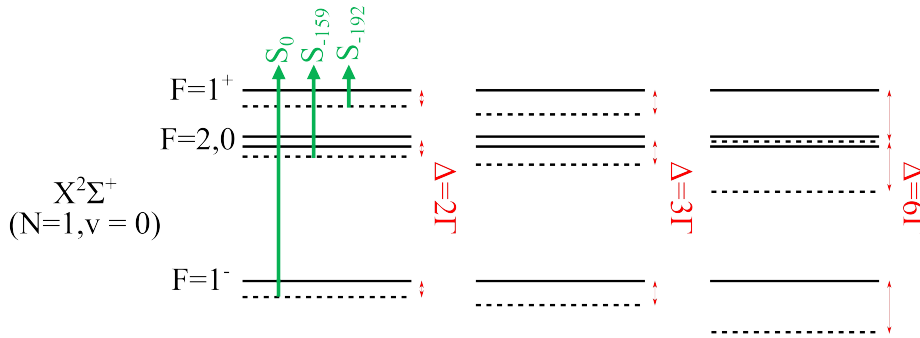


Figure 4.7: Energy level diagram (not to scale) of the YbF electronic ground state, ($\nu = 0$, $N = 1$). Dashed lines depict the blue detuning of the cooling light. All three laser frequencies, S_0 , S_{-159} and S_{-192} are detuned by the same amount, Δ . The detuning increases left to right with the final detuning of $\Delta = 6\Gamma$ resulting in S_{-192} being approximately resonant with the $X^2\Sigma^+(F = 2,0) \rightarrow A^2\Pi_{1/2}$ transitions.

It was hypothesised that the red-detuned excitations could be somewhat mitigated by changing the balance of laser power allotted to each sideband. Therefore, several investigations were undertaken to test empirically what balance of sideband powers would be optimal [1]. Table 4.1 shows the results of five experimental configurations for the cooling sidebands. Sisyphus cooling was the mechanism attempted in all cases, and the magnetic field was set to 0.5 G.

Configurations one and two both used only one frequency component. Cooling was inefficient

Configuration	r_0	r_{-159}	r_{-192}	Δ (Γ)	S_{1D}
1	0	1	0	6	0
2	1	0	0	3	0.1
3	0.33	0.33	0.33	3	0.2
4	0.96	0.02	0.02	6	0.5
5	0.5	0.5	0	3	1.2

Table 4.1: Results of five different cooling light configurations. The total laser power was split between the three sidebands. A fraction, r_δ was allotted to each sideband S_δ . The detuning, Δ , and a relative measure of the number of ultracold molecules, S_{1D} , are provided for each configuration.

because the single frequency was far from resonance from the other two transitions. Only the $F = 2, 0$ components were addressed in the first configuration. Even with $\Delta = 6\Gamma$, the cooling light was too far detuned from $F = 1^-$ and $F = 1^+$, resulting in low photon scattering rates.

In configuration 3, the laser power was split evenly between the three sidebands. The detuning, $\Delta = 3\Gamma$, of this configuration is depicted in the centre energy level diagram of figure 4.7, where S_{-192} is equally blue and red detuned from the $F = 1^+$ and $F = 2, 0$ states respectively. This resulted in poor cooling because of the red-detuned scattering. However, it was better than the first two configurations because all transitions were addressed.

The fourth configuration cooled more than double the number of molecules in configuration three. S_0 was allocated 96% of the laser power, whilst S_{-159} and S_{-192} received 2% each. The optimal detuning found was $\Delta = 6\Gamma$ and is depicted in the right-most energy level diagram of figure 4.7. All levels were addressed by blue-detuned light, but the $F = 1^+$ and $F = 2, 0$ states had low scattering rates due to the low power of the laser light addressing them. In this configuration, cooling was mainly from $F = 1^-$ while the other components mainly act as repumpers. However, the large blue detuning needed to optimise the configuration resulted in the S_{-192} component becoming approximately resonant with the S_{-159} transition; this was detrimental to cooling.

Configuration five was the best of the configurations - providing the most laser-cooled molecules. The cooling laser power was split evenly between the S_0 and S_{-159} . The cooling was much more effective in the absence of S_{-192} because there was no longer any red-detuned light. The high laser power and relatively small blue-detuning of S_{-159} provided enough off-resonant scattering from the $F = 1^+$ state to prevent it from becoming dark to the cooling light. Since it was so effective, this

configuration was used for the rest of the measurements presented in this chapter.

4.3.2 Cooling light detuning

Figure 4.8 shows the variation of S_{1D} as a function of cooling light detuning. The experiment employed a total cooling light power equivalent to $450 I_{\text{sat}}$ and a magnetic field strength of $B = 0.5$ G. The results demonstrate that as the detuning increased, the number of laser-cooled molecules also increased, reaching an optimum around $\Delta = 5 \Gamma$. However, beyond this point, the effectiveness of cooling diminished. This observation aligns with the anticipated behaviour of the AC Stark shift, which initially starts small at resonance, increases to a maximum, and then decreases. Alternatively, this trend could be attributed to molecules decaying to the $F = 1^+$ state and failing to scatter sufficient photons from the S_{-159} laser when the detuning becomes significant.

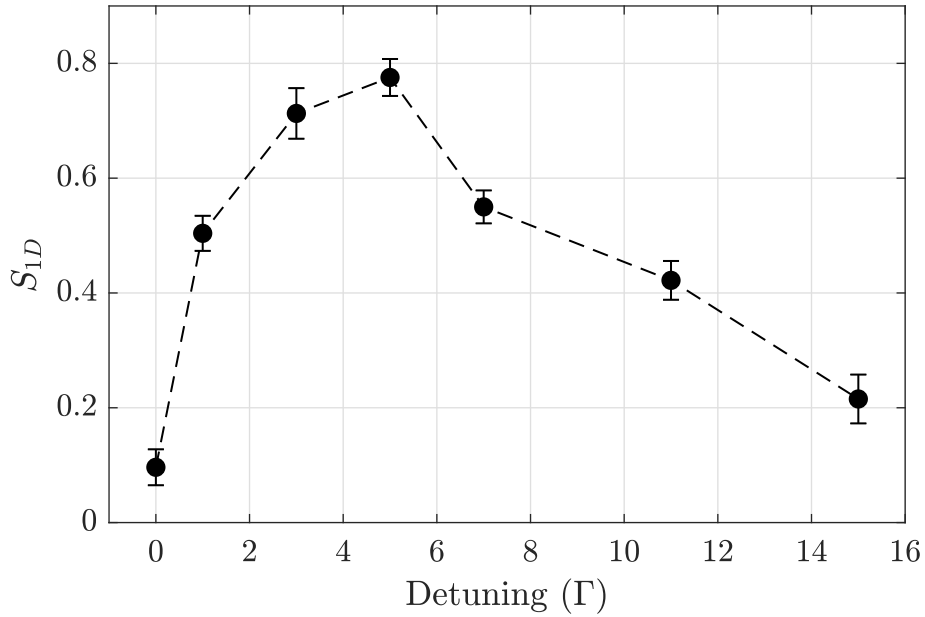


Figure 4.8: S_{1D} as a function of detuning, Δ . A magnetic field strength of $B = 0.5$ G was used, and a maximum cooling light intensity of $450 I_{\text{sat}}$.

4.4 Experimental results: two-dimensional laser cooling

Once satisfied that one-dimensional laser cooling was optimized in each transverse axis, we proceeded to two-dimensional laser cooling. Measurements were made in the same manner as for

the one-dimensional cooling measurements but with the addition of CCD1 for measuring temperature. In this section, I present results for two sub-Doppler cooling mechanisms and characterize the 2D laser-cooled molecular beam in terms of its temperature and the number of ultracold molecules.

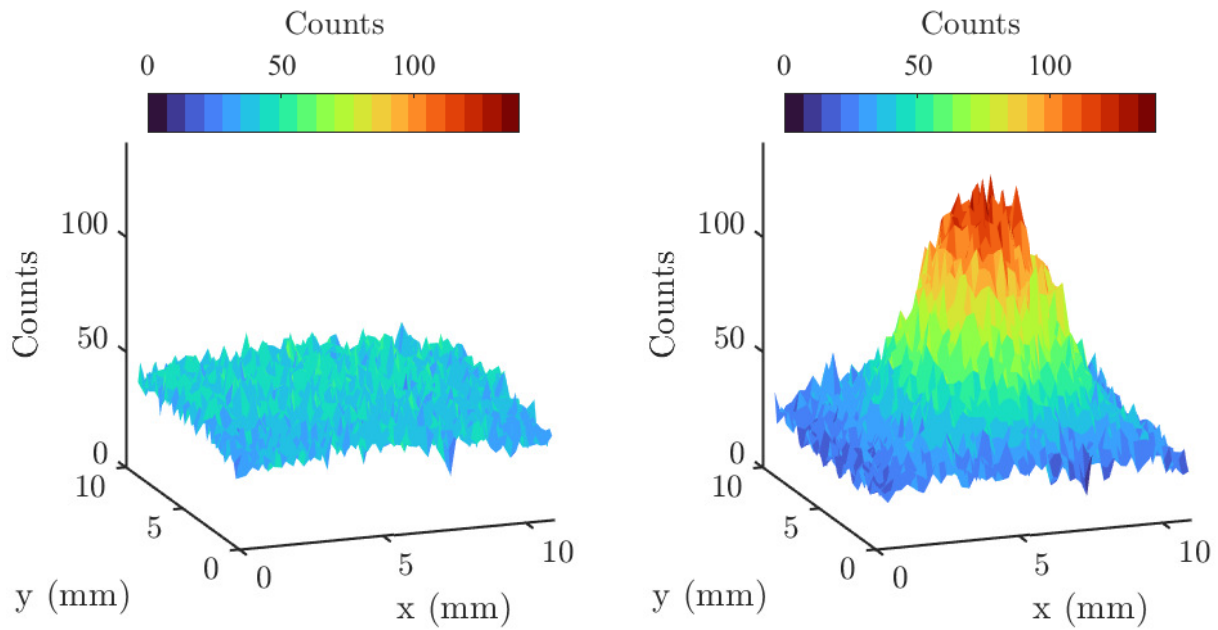
Without cooling applied, the molecule beam diverged rapidly and had a very broad 2D Gaussian profile by the time it reached CCD2⁴. This is seen in the example data in figure 4.9a, where the CCD counts were nearly flat in the xy -plane. Figure 4.9b shows the 2D laser-cooled molecules from the same measurement. A narrower 2D Gaussian profile was observed, with a larger amplitude; these were the ultracold molecules.

The ratio of the cooled and uncooled photos (figures 4.9b and 4.9a) is shown by a grey mesh in figure 4.9c, and is also inset as an image. A 2D Gaussian $g(x, y) = \frac{A}{2\pi} e^{-(x-x_0)^2/(2\sigma_x^2)} e^{-(y-y_0)^2/(2\sigma_y^2)} + b$ was fitted to the data and is shown as a coloured surface. From fits such as this, the width of the molecular beam is extracted for temperature measurements. The number of molecules is calculated using the quantity $S_{2D} = A\sigma_x\sigma_y/L^2$, where $A\sigma_x\sigma_y$ is the volume under a 2D Gaussian⁵. However, partially cooled molecules are seen outside the 2D Gaussian peak; they are easier to see in the inset image, forming a cross which connects the image's corners. These molecules cause the goodness-of-fit to be lower than otherwise acceptable, with an adjusted R-squared value of 0.88. Nonetheless, it was found that for several measurements, the integral of the ratio data was within 2.5% of the fit volume and within the 95% confidence bound of the fit. Similarly, estimates of distribution widths agree well with 2D Gaussian fits. Thus whilst the goodness-of-fit was smaller than desired, the model is suitable for this analysis.

In the following sections, several properties of the laser-cooled molecular beam are characterised using S_{2D} as the figure of merit. In addition, temperature measurements of the molecular beam are presented (sec. 4.4.4), leading to calculations of the brightness of the beam (sec. 4.4.5) and improved sensitivity of an eEDM measurement (sec. 4.4.6).

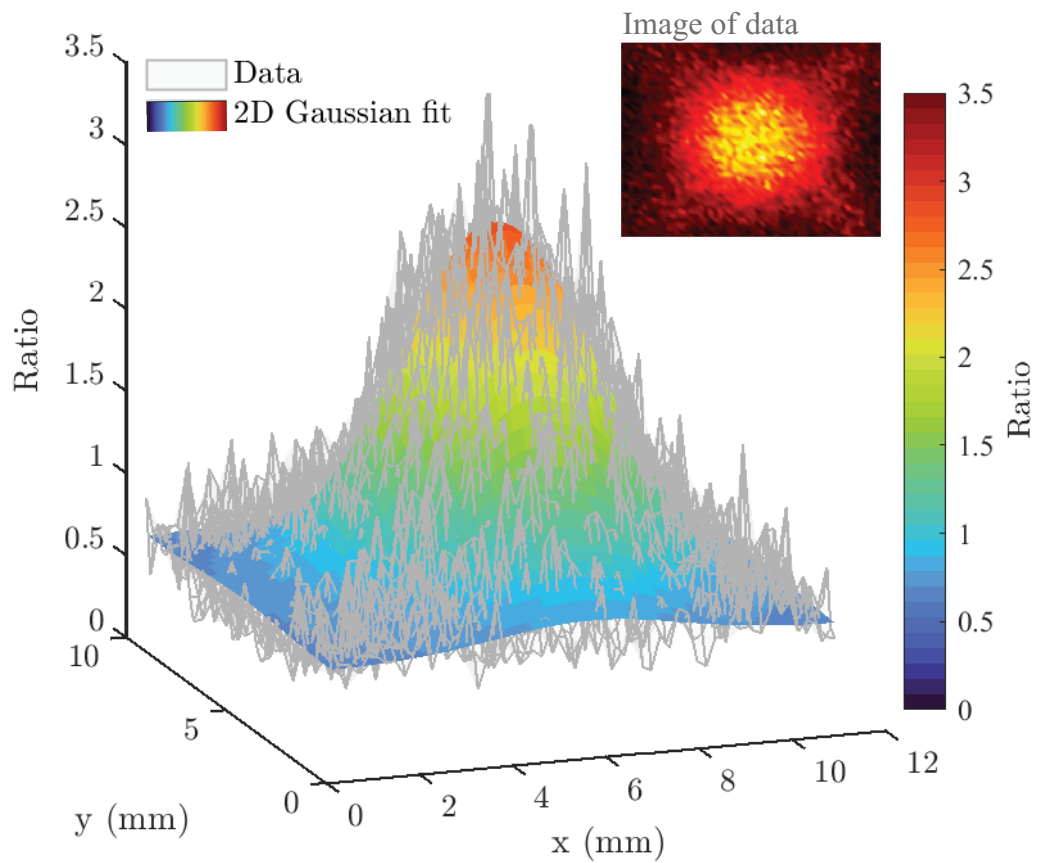
⁴Whilst the uncooled molecular spatial distribution appeared to be flat at CCD2, its Gaussian profile was observed at CCD1 (see figure 4.6b).

⁵ S_{2D} is proportional to the number of ultracold molecules and is analogous to S_{1D} in section 4.3.



(a) Uncooled beam.

(b) Cooled beam.



(c) Ratio of cooled to uncooled counts in grey (also inset), with a colour scaled 2D Gaussian fit.

Figure 4.9: Examples of 2D laser cooling measurements taken using CCD2. The photos were background subtracted and averaged to produce two images: molecules with and without 2D laser cooling applied. A two-dimensional Gaussian is fitted to the ratio of the two images.

4.4.1 Comparison of sub-Doppler cooling mechanisms

Figure 4.10 shows S_{2D} as a function of the applied magnetic field strength for the molecules. The black data correspond to the $XY_{\parallel}$ configuration, where the counter-propagating lasers have parallel linear polarisations. In contrast, the blue data represent the $XY_{\perp}$ laser cooling configuration, where the counter-propagating cooling lasers have linear polarisations that are perpendicular to each other. As discussed in section 2.3.4, these distinct configurations exhibit different dependencies on the applied magnetic field. This measurement can be used to identify any imperfections in the beam polarisations or the applied magnetic field by comparing the behaviour of the two sub-Doppler cooling mechanisms.

In the $XY_{\parallel}$ configuration, a minimum is observed at $B = 0$. This result aligns with expectations since this sub-Doppler cooling mechanism relies on the presence of a magnetic field to retain molecules in the cooling cycle. However, even at $B = 0$, molecules were still undergoing cooling, as evidenced by $S_{2D} > 3$. Rigorous calibration of the magnetic fields in the cooling region was conducted, and the shim coils were used to eliminate background fields. Therefore, the unexpected cooling was attributed to the polarisation of the cooling beams. Analysis of the beam properties revealed that their polarisations exhibited variations as they propagated through the cooling region. This phenomenon's underlying cause was stress-induced birefringence in the windows of the panoramic vacuum chamber. Each time a beam passed through a window, its polarisation would experience a rotation. The resulting lattice formed by the beams comprised both intensity and polarisation gradients. To address this issue, measures were taken to reduce stress in the windows, such as reseating them and ensuring that the bolts connecting the flange to the chamber were not over-tightened.

The data from the $XY_{\perp}$ configuration exhibit a weak dependence on the magnetic field strength within the range $|B| < 1$ G. If the polarisations of the cooling beams were consistently perpendicular throughout the cooling region, we would expect maximum cooling at the magnetic field minimum. However, due to imperfections introduced by the vacuum windows, both polarisation and intensity gradients were present.

Simulations have predicted that the $XY_{\perp}$ configuration should yield better cooling results compared to the $XY_{\parallel}$ configuration [1]. The observed discrepancy between the results and simulations could be attributed to the imperfections in beam polarisation. Surprisingly, the $XY_{\parallel}$ configuration resulted in a higher number of cooled molecules compared to the $XY_{\perp}$ configuration when the magnetic field exceeded 0.5 G. Because of this, the rest of the measurements presented in the chapter all use the $XY_{\parallel}$ configuration.

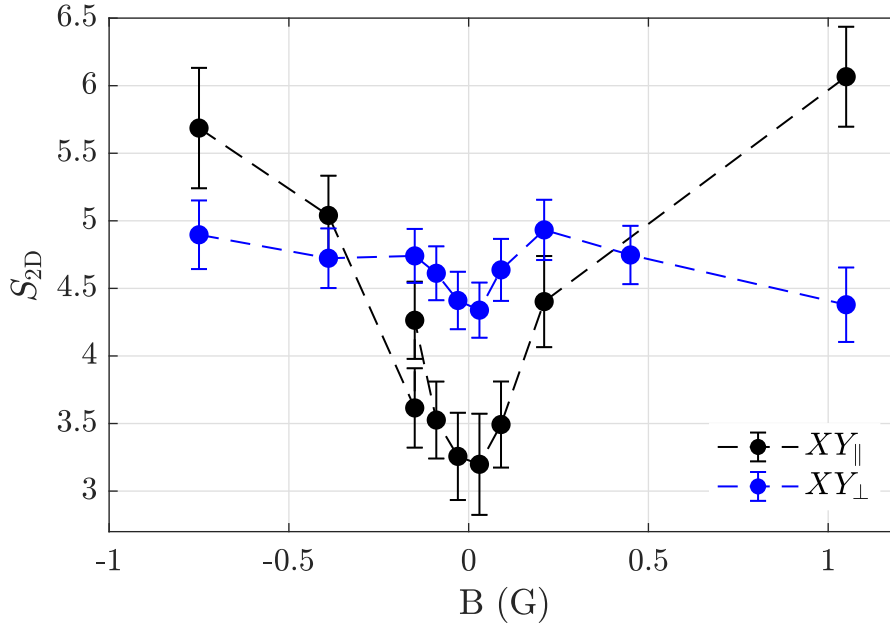


Figure 4.10: S_{2D} as a function of applied magnetic field strength, B . Blue circles show the $XY_{\perp}$ polarisation configuration, whilst black circles show the $XY_{\parallel}$ configuration.

4.4.2 Cooling light power and detuning

Figure 4.11 illustrates the variation of S_{2D} with cooling light detuning. Three different data sets are presented, each corresponding to different peak intensities of the cooling laser. As the intensity of the cooling light increased, the number of cooled molecules also increased. This can be attributed to the larger AC Stark shifts induced in the molecules, resulting in more kinetic energy loss per scattered photon. Notably, we did not observe saturation of this effect within the intensity limits imposed by our laser system.

Furthermore, the optimal detuning for each curve increased with the peak intensity of the cooling light. This observation aligns with the behaviour of the AC Stark shift, as higher intensities require

larger detunings to maximise the effect. Additionally, increased detuning reduced the excitation rate, allowing each molecule more time to reach the top of a potential peak before scattering a photon.

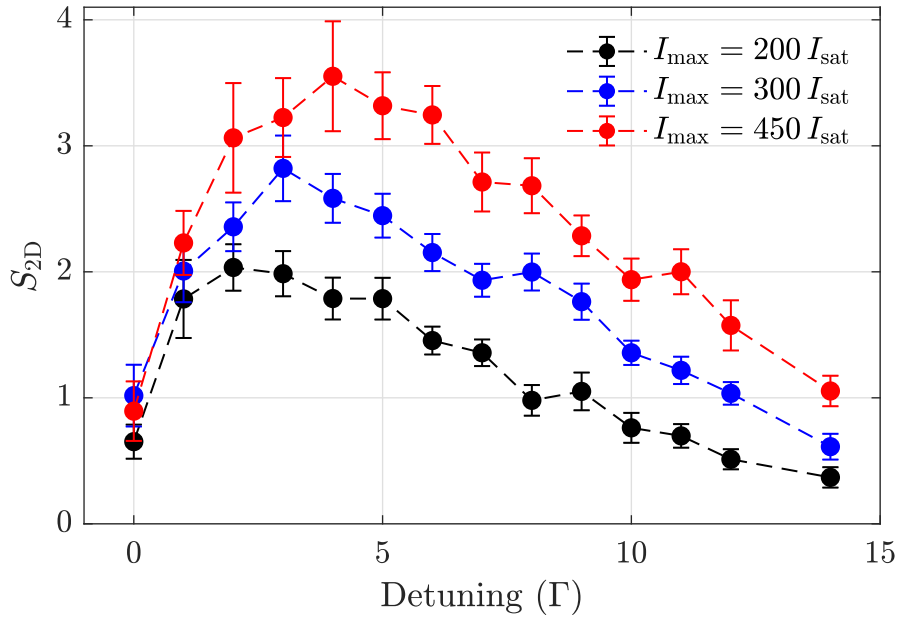


Figure 4.11: S_{2D} as a function of cooling light detuning. The three sets of data correspond to different cooling light intensities.

4.4.3 Vibrational repump powers

Figure 4.12 shows the relationship between S_{2D} and the laser power used for the first vibrational repump. The measurements in the figure were conducted with the same cooling light detuning 6Γ , but using three different intensities of cooling light. The second vibrational repump laser was operated at its maximum power output.

All three measurements exhibit a similar trend. As the power of the repump laser increased, the number of ultracold molecules also increased. However, this effect quickly reached saturation, indicating that we had adequate laser power to repump molecules from the first excited vibrational state. Additionally, as the intensity of the cooling light increased, we anticipated observing an increase in the saturation power. This expectation arose from the fact that the scattering rate of the cooling transition rises with intensity, causing molecules to exit the cooling cycle more frequently and decay to higher vibrational states. Consequently, more repump laser power was necessary to

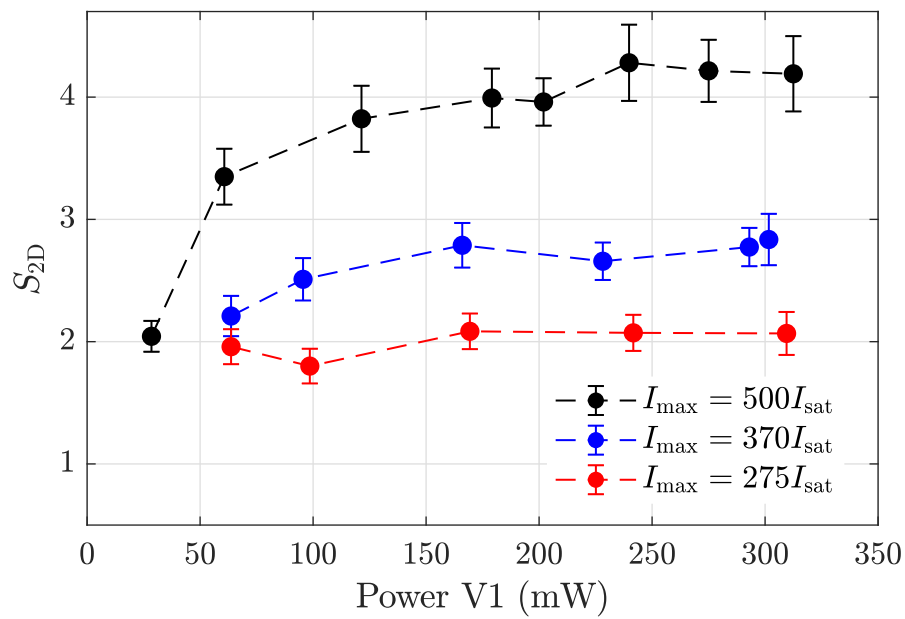


Figure 4.12: S_{2D} as a function of laser power used for the first vibrational repump. A cooling light detuning of 6Γ was used, and three different cooling light intensities were tried.

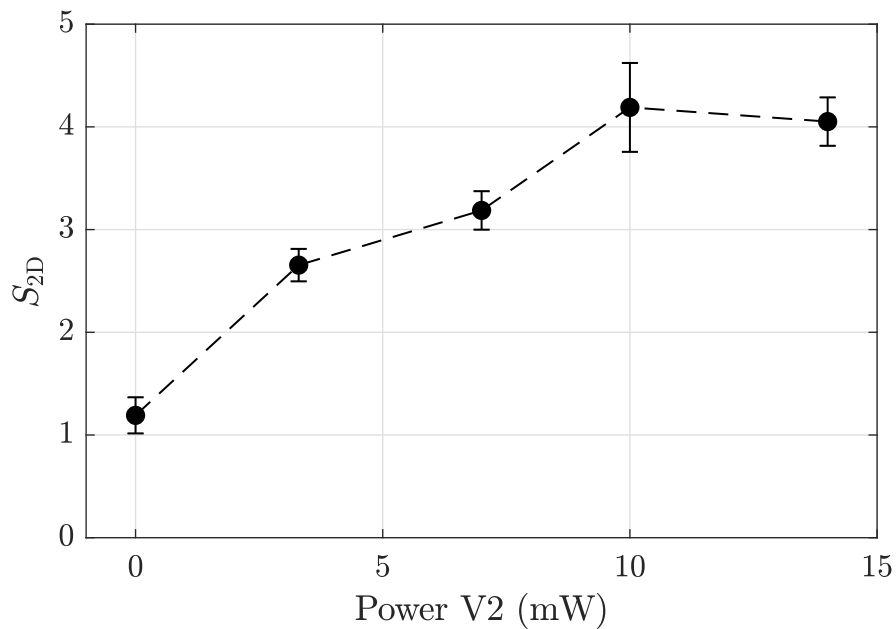


Figure 4.13: S_{2D} as a function of laser power used for the second vibrational repump. Cooling light with peak intensity $I_{\max} = 500 I_{\text{sat}}$ and detuning of 6Γ was used, and the first excited vibrational repump laser operated at maximum power.

return the molecules to the cooling cycle.

Figure 4.13 shows S_{2D} as a function of the laser power used to address the second excited vibrational state. For this measurement, the cooling light detuning was kept constant at 6Γ and both the cooling laser and first excited vibrational repump laser operated at their maximum power outputs. The data show a similar trend as those shown in figure 4.12. As the repump laser power increased, so did the number of ultracold molecules. The increasing trend appeared to saturate above 10 mW, but unfortunately, the laser system's output limited the repump laser power.

4.4.4 Molecular beam temperature measurements

Characterisation of the laser-cooled molecular beam has, until this section, been focused on the number of ultracold molecules; this being important for reducing the statistical uncertainty of an eEDM measurement. However, the temperature of the molecular beam is just as important for us as this collimation of the beam will allow the molecules to propagate through the interaction region for an extended period of time. To measure the temperature of the freely propagating laser-cooled molecules, their transverse rms width, $\sigma_{x,y}$, must be monitored at different moments in time to observe their expansion. The two cameras, CCD1 and CCD2, facilitate this but only provide temperature measurements of the beam in the y-axis because CCD1 views the molecular beam in the yz-plane. We assert that because the x and y-axis rms widths are similar at CCD2, extending our conclusions to the x-axis temperature is fair. The temperature, T , of the beam in the y-axis is calculated as follows, [1];

$$T = \frac{m}{k_B} v_z^2 \frac{\sigma_{y_2}^2 - \sigma_{y_1}^2}{z_2^2 - z_1^2}, \quad (4.1)$$

where m is the mass of a YbF molecule, k_B is Boltzmann's constant, v_z is the average longitudinal velocity of the beam, σ_{y_i} is the rms width of the beam in the y-axis at position i and z_i is the longitudinal position where y_i was measured. For the measurements presented in this section, the

average forward velocity of the molecular beam was 200 m s^{-1} (except when helium flow was varied, which made the forward velocity a dependent variable). The two detectors were positioned at $z_1 = 0.9 \text{ m}$ and $z_2 = 1.5 \text{ m}$, giving the molecules 3 ms of free propagation time between measurements.

Figure 4.14 shows the molecular temperature as a function of cooling light detuning. The two data sets were taken with different peak cooling light intensities whilst a magnetic field of strength 0.5 G was being applied. The blue crosses show data taken using $I_{\text{max}} = 240 I_{\text{sat}}$. As the detuning increases, the temperature of the molecules decreases until a minimum is reached around 9Γ . The measurement was resolution limited in this region, such that the molecular temperature was less than one standard deviation above 0 K. Of course, the molecules cannot have a temperature less than 0 K, but we can say with 95% confidence that the temperature is less than $220 \mu\text{K}$. As the detuning increased beyond the minimum, the temperature increased. This happened because the photon scattering rate decreased, making cooling less effective.

The black circles in figure 4.14 show data taken with the cooling light at maximum intensity⁶. As the detuning increased, the temperature of the molecules would decrease. However, unlike in the lower intensity case, no minimum was observed in the range of detunings 1 – 15Γ . Temperature measurements at higher detunings were challenging because of the small number of cooled molecules (see figure 4.11). When the ultracold molecular signal became small relative to the uncooled molecules, the ratio of the signals would be near flat, and noise would disrupt the fitting algorithms used during analysis. However, the trend was clear; with higher-intensity cooling light, larger detunings were needed to reduce the temperature of the molecular beam.

Comparing the two data sets in figure 4.14, we see that, for the same detuning, the higher intensity cooling light produced a molecular beam with a higher temperature. We also see in figure 4.11 that higher intensities produced more cooled molecules. These results were linked by the higher laser cooling capture velocity when the cooling light intensity was increased; more molecules were laser-cooled, but not all were cooled long enough to reach their equilibrium temperature. Whilst

⁶This was the maximum power output of the cooling laser at the time of the measurement. Other measurements in this chapter show higher cooling light intensities because the power output of the laser varied a little over time due to slow drifts in the setup. These were drifts in the optimal temperature of the laser doubling crystal and the fibre coupling between the seed laser and amplifier.

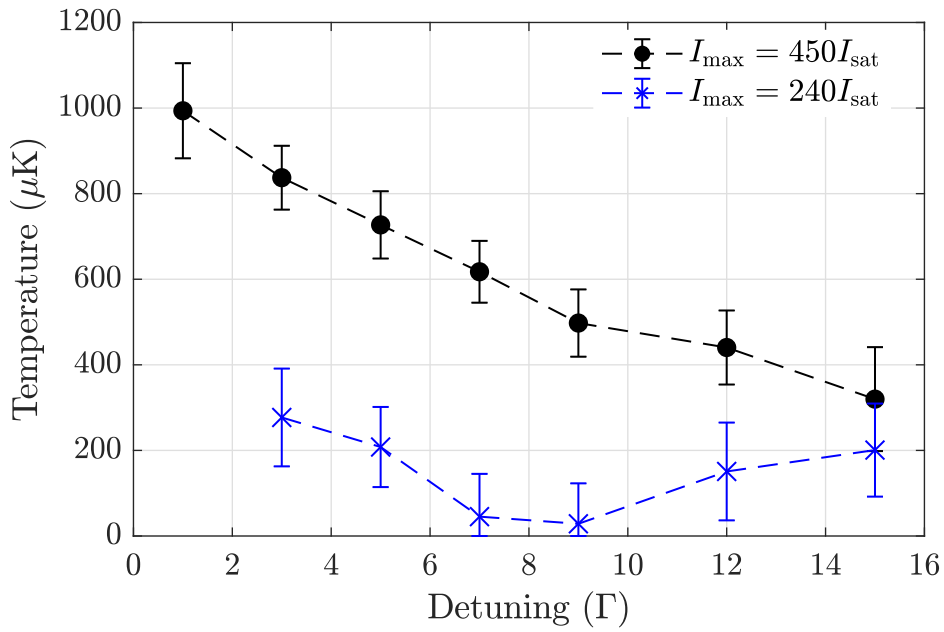


Figure 4.14: Transverse temperature of the molecular beam as a function of cooling light detuning. Two different cooling light intensities were tried, and a magnetic field of 0.5 G was applied.

the partially cooled molecules had low transverse velocities, they were not as slow as those which reached equilibrium, and they broadened the spatial distribution of the beam. Therefore, these temperature measurements were likely higher than the equilibrium temperature. Simulations indicated that the laser cooling critical velocity was far smaller than the capture velocity at these intensities of cooling light and that the equilibrium temperature could have been as low as $10 \mu\text{K}$ [1]. All of our temperature measurements observed this link between maximising the number of ultracold molecules and increased temperature.

Figure 4.15 shows the temperature of the molecular beam as a function of the strength of the applied magnetic field. The beam's temperature is minimised by minimising the applied magnetic field strength. However, this comes with the cost of having very few ultracold molecules. The trend is similar to that of figure 4.10, where the number of ultracold molecules increases with the strength of the applied magnetic field. This is consistent with the explanation provided for figure 4.14 –the temperature of the molecules increases with the number of ultracold molecules.

Figure 4.16 shows the effect of varying helium flow rates in the molecular source. The black circles show the temperature of the molecular beam, whilst the blue crosses show the forward speed of the beam. As the beam became faster, the molecules spent less time being cooled, and the temper-

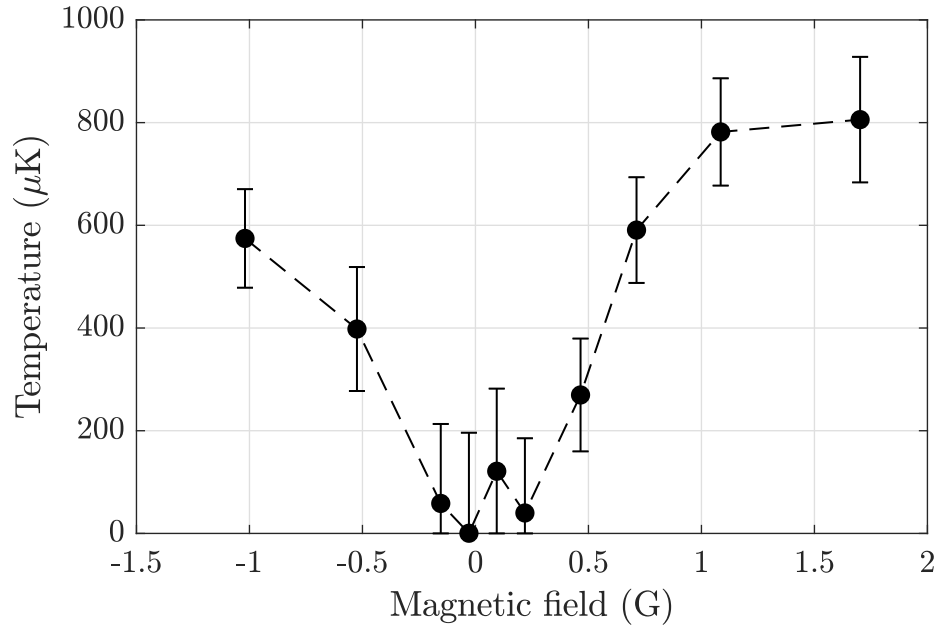


Figure 4.15: Transverse temperature of the molecular beam as a function of the applied magnetic field strength. The cooling light had intensity $450 I_{\text{sat}}$ and a detuning of 7Γ .

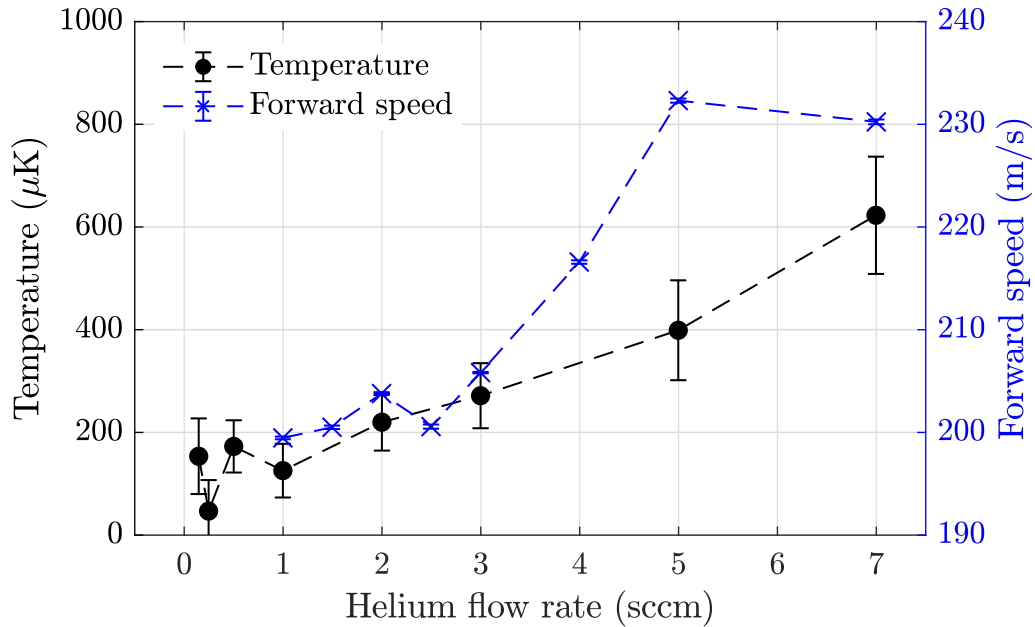


Figure 4.16: Transverse temperature (black circles) and forward speed (blue crosses) of the molecular beam as a function of the helium flow rate in the molecular source. The applied magnetic field strength for these measurements was 0.5 G, whilst the cooling light had intensity $450 I_{\text{sat}}$ and a detuning of 7Γ .

ature of the ultracold molecules increased. However, this did not account for the molecular beam's flow or divergence variation. This was not measured as we were driven by empirical optimisation of the setup. Nevertheless, we found that the optimal helium flow rate⁷ was between 1 and 2 SCCM - providing a slow but intense beam of ultracold molecules.

4.4.5 Brightness of the laser cooled molecular beam

For precision measurement of the eEDM, we require a beam with many molecules, high collimation and a small area. A good measure for this is a beam's brightness, or radiance, which measures the flux per unit area and solid angle. The radiance, $\mathcal{R}$, of our molecular beam is defined as follows [1, 104],

$$\mathcal{R} = \frac{\dot{N}}{\Delta x^2 \Delta \Omega}, \quad (4.2)$$

where $\dot{N}$ is the number of molecules per unit time passing through a transverse area Δx^2 with solid angle $\Delta \Omega$ [1]. The solid angle can be described in terms of molecular velocities,

$$\Delta \Omega = \pi (\Delta v_{\perp} / v_z)^2, \quad (4.3)$$

where the transverse temperature, T , defines the range of transverse velocities $\Delta v_{\perp} = \sqrt{k_B T / m}$ [1].

Under typical operating conditions, the molecular beam consists of approximately 2×10^5 ultracold molecules per bunch. Running the molecular source at a frequency of 10 Hz yields a production rate of $\dot{N} \approx 2 \times 10^6$ ultracold molecules per second. These molecules occupy an area of approximately $5 \times 5 \text{ mm}^2$ and exhibit a minimum temperature of $T < 200 \mu\text{K}$. These parameters result

⁷SCCM is a measurement of flow rate and stands for Standard Cubic Centimeters per Minute. It is a unit used to measure the flow rate of gases, typically under standard conditions of temperature and pressure (0°C and 1 atm).

in a brightness of $\mathcal{R} > 1 \times 10^{17}$ molecules/m²/sr/s. This exceeds the brightness of the uncooled molecular beam by at least two orders of magnitude.

4.4.6 Impact for an eEDM measurement

In contemplating the advancements in the measurement of the eEDM, it is crucial to address the effects of quantum projection noise on the precision of future experiments [1, 3, 105]. During the detection phase of an eEDM experiment, where the populations of quantum states are measured, the quantum states of the system undergo a projection onto a set of measurement eigenstates. This process introduces quantum projection noise as a fundamental limit to the measurement's precision. At the quantum projection noise limit, the statistical uncertainty, σ_{d_e} , in a measurement of the eEDM is,

$$\sigma_{d_e} = \frac{1}{C} \frac{\hbar}{E_{\text{eff}} \tau} \frac{1}{2\sqrt{N_{\text{mol}}}} \quad (4.4)$$

where C is the measurement contrast, a parameter that captures the signal-to-noise ratio; $\hbar$ is the reduced Planck constant, encapsulating the quantum nature of the measurement; E_{eff} is the effective electric field experienced by the molecules, which enhances the sensitivity to the eEDM; τ is the interrogation time, representing the duration over which the system evolves coherently and the eEDM interaction can accumulate phase; and N_{mol} is the number of molecules detected, with statistical uncertainty scaling inversely with the square root of this number [1, 3, 105].

Taking into account the realistic constraints of our apparatus and the molecular beam's velocity, an interrogation time of 25 ms is achievable. With an anticipated electric field of 20 kV/cm within the interaction zone, this yields an effective field strength of 17.5 GV/cm. Drawing on typical values from prior YbF experiments—contrast C of 0.9, detection efficiency of 30%, and a duty cycle of 0.5—we arrive at a daily statistical uncertainty of $\sigma_{d_e} = 5 \times 10^{-30}$ ecm. Over the course of one hundred days, leveraging the square root law for statistical improvement, this precision improves

to $\sigma_{d_e} = 5 \times 10^{-31}$ e cm. Such a result would be 36 times more sensitive than measurements made using a supersonic YbF source without laser cooling and would be the most precise measurement of the eEDM ever made [1, 3, 16].

4.5 Summary

This chapter presents the results of one-dimensional and two-dimensional transverse laser cooling experiments conducted on a YbF molecular beam. Firstly, after several upgrades to the apparatus, one-dimensional cooling was optimised. A new sideband power configuration was identified with our high-intensity cooling light. This new configuration relied on the absence of the sideband addressing $F = 1^+$ of the cooling ground state to efficiently cool molecules.

Results were then presented for two-dimensional transverse laser cooling of the molecular beam. Two sub-Doppler laser cooling regimes were applied to the molecular beam. We found it challenging to preserve the polarisation of the cooling beams but still managed to demonstrate the expected trends in molecule number versus applied magnetic field strength. It was then shown that further increases in laser power could lead to greater numbers of laser-cooled molecules, even in the experiment's high-intensity regime. Similar measurements showed that the cooling scheme is not limited by the intensity of vibrational repump laser light.

The temperature of the molecular beam was then characterised, with the best measurement finding a temperature $< 200 \mu\text{K}$ with 95% confidence. The close link between the molecular temperature and the number of ultracold molecules was discussed in the context of the capture and critical velocities of the cooling scheme. The brightness of the molecular beam was calculated to be $> 1 \times 10^{17}$ molecules/m²/sr/s - surpassing the brightness of the uncooled molecular beam by over two orders of magnitude.

Finally, the sensitivity of an electric dipole moment (eEDM) measurement using this laser-cooled beam was estimated. After one hundred days of measurement, the projected sensitivity was $\sigma_{d_e} = 5 \times 10^{-31}$ e cm, representing a 36-fold improvement compared to measurements using a supersonic YbF source.

Chapter 5

YbF molecular funnel

To generate a low transverse velocity atomic or molecular beam, cooling techniques such as laser cooling are employed. Although laser cooling significantly reduces the beam's thermal expansion, losses occur due to inadequate matching between the beam's phase space distribution and the phase space acceptance of the laser cooling regime. To address this issue, a concept known as the "atomic funnel" was introduced at Stanford University in 1990 [106]. This technique involved magnetically guiding sodium atoms to cooling lasers using a quadrupole magnetic field, effectively enhancing the beam's brightness and reducing the transverse velocities of the atoms. The combination of magnetic lensing followed by collimation with cooling lasers became known as funnelling due to the trajectory that the particles trace out [107].

As discussed in chapter 4, laser cooling of molecules has proven to be more challenging than atoms. The presence of rotational and vibrational states complicates the identification of closed cooling cycles. Consequently, only a limited number of molecular species have been successfully laser-cooled thus far, with YbF being one of them. However, the concept of a molecular funnel has yet to be demonstrated. Implementing a molecular funnel in an electron electric dipole moment measurement would significantly increase the number of available molecules, thereby reducing the statistical uncertainty associated with the measurement. Given our successful demonstration of transverse sub-Doppler laser cooling of the YbF beam, we are now poised to implement a molecular funnel for the first time.

This chapter focuses on the simulations and design of a magnetic lens for funnelling YbF molecules. The assembly of the lens has been completed, and it is currently awaiting installation in the experimental apparatus.

5.1 Magnetic lensing

5.1.1 Magnetic force on a neutral atom or molecule

When an electrically charged particle moves perpendicular to a uniform magnetic field, it experiences the Lorentz force. However, the Lorentz force does not apply to neutral atoms or molecules. Instead, they can experience another type of force in the presence of magnetic field gradients. This force arises from the Zeeman effect, which is described in section 2.2.1. The Zeeman energy shift varies with the strength of the magnetic field, so when there are gradients in the field, neutral particles can experience a force. Figure 2.4 illustrates the energy shifts of the YbF magnetic sublevels as a function of magnetic field strength.

5.1.2 Manipulating neutral particle beams with magnetic fields

Halbach arrays

The concept of a Halbach array involves arranging permanent magnets in a row with varying magnetisation directions, resulting in a strong magnetic field on one side and near zero on the other. While the invention is commonly associated with Klaus Halbach, the effect was actually demonstrated earlier, in 1970, by J. Winey during the development of an electromagnetic transducer [108, 109]. J. Mallinson provided the first detailed description of the magnetisation pattern in 1973, where it was proposed as an improvement for tape recording devices [110]. Since then, Halbach arrays have found applications in various devices, ranging from everyday items like fridge magnets to advanced magnetic levitation systems for trains [111].

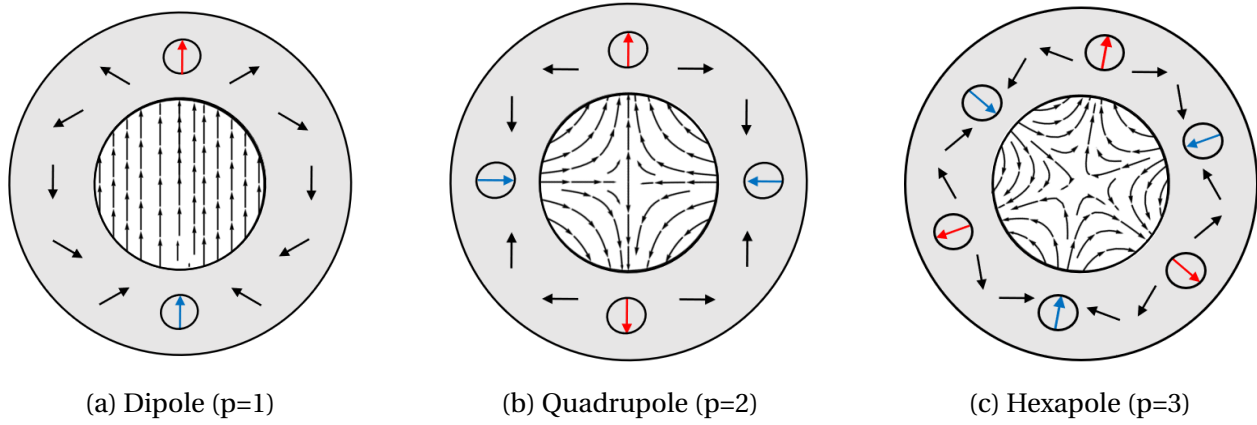


Figure 5.1: Cross section of three ideal Halbach cylinders with different pole numbers. Magnetisation direction is illustrated using arrows in the magnetic material (shaded grey) and varies continuously around each cylinder. Circled arrows guide the reader to the pairs of poles. The magnetic vector field is drawn in the bore of the cylinder.

Halbach cylinders

A Halbach array can be curled around to form a cylinder, known as a Halbach cylinder [108]. In the ideal case, the cylinder is infinitely long, and the magnetisation direction varies continuously around the cylindrical magnet. Figure 5.1 illustrates cross-sections of three ideal Halbach cylinders. The magnetisation direction, indicated by arrows, rotates continuously, resulting in one, two, or three poles pointing radially outwards (circled red arrows) and the same number pointing radially inwards (circled blue arrows). The magnetisation variation is confined to the plane shown in the figure. In cylindrical coordinates, the magnetization $\vec{M}$ depends on the azimuth ϕ , the number of pole-pairs p , and the magnetic remanence M_r :

$$\vec{M}(\rho, \phi, z) = \vec{M}(\phi) = M_r \left(\cos \left(p \left(\phi - \frac{\pi}{2} \right) \right) \hat{\rho} + \sin \left(p \left(\phi - \frac{\pi}{2} \right) \right) \hat{\phi} \right). \quad (5.1)$$

Figure 5.1 also demonstrates that the magnetic field lines are completely confined to the bore of each cylinder, with zero field outside.

However, it is impossible to manufacture a single magnet with continuously varying magnetisation directions, so any physical realisation of a Halbach cylinder must approximate it by segmenting

the magnet. This approximation introduces higher-order poles to the field, which can lead to undesirable effects. Additionally, non-ideal Halbach cylinders have finite lengths, introducing end effects where some magnetic flux "escapes" the cylinder [108, 112, 113].

In 1980, Halbach derived the expression for the norm of the magnetic flux density in the bore of a segmented but infinitely long cylinder [108, 112, 113]:

$$|\vec{B}(\rho, \phi)| = B_p \left(\frac{\rho}{\rho_1} \right)^{p-1}. \quad (5.2)$$

Here, ρ_1 represents the bore radius of the cylinder, and B_p denotes the strength of the flux density at the inner surface of a cylinder with p pole-pairs.

Dipole Halbach cylinder

Figure 5.1a depicts a cross-section of an ideal dipole Halbach cylinder. In this configuration, the magnetic field within the bore is homogeneous, resulting in the absence of magnetic field gradients. Consequently, a neutral particle experiences a constant Zeeman energy shift throughout the bore, leading to no force acting on it.

However, a charged particle travelling along the bore will encounter a force. Thus, this device finds application in deflecting charged particle beams. It is widely utilised in the field of particle physics for tasks such as particle injection and ejection from accelerators [114].

Quadrupole Halbach cylinder

Figure 5.1b illustrates a cross-section of an ideal quadrupole Halbach cylinder. In this ideal scenario, the magnetic field has no axial components and is inhomogeneous in the transverse plane of the cylinder. A neutral atom or molecule that has a linear Zeeman shift will feel a constant force directed towards the centre of the cylinder.

On the other hand, charged particles exhibit different behaviour. They are focused in one axis within the quadrupole cylinder. A pair of sequential quadrupole cylinders can be employed to focus in both transverse axes, with the cylinders rotated by $\pi/2$ relative to each other [115]. This technique is commonly employed in particle accelerators for effective beam focusing [115].

Hexapole Halbach cylinder

Figure 5.1c illustrates a cross-section of an ideal hexapole Halbach cylinder. In this ideal scenario, the magnetic field within the cylinder has no axial components and exhibits inhomogeneity in the transverse plane. The norm of the magnetic flux density, denoted as $|\vec{B}(\rho, \phi)|$, varies quadratically with respect to the radial distance from the centre of the cylinder. By employing equation 5.2, we can express the magnetic flux norm for a segmented hexapole cylinder as:

$$|\vec{B}(\rho, \phi)| = B_3 \left(\frac{\rho}{\rho_1} \right)^2 \quad (5.3)$$

From Kaenders et al. (1996) [113], B_3 is given as,

$$B_3 = \frac{3}{2} B_r \left(1 - \left(\frac{\rho_1}{\rho_2} \right)^3 \right) G_{s,3}, \quad (5.4)$$

where B_r is the magnetic remanence of the magnets, ρ_2 is the outer radius of the cylinder and $G_{s,3}$ is the geometry factor which accounts for the segmentation of the cylinder. From Halbach (1980) [108] and Kaenders et al. (1996) [113] we have a general form for the geometry factor, $G_{s,p}$, which depends on the number of magnet segments, s , and the number of pole-pairs, p :

$$G_{s,p} = \frac{\sin((p+1)\pi/s)}{(p+1)\pi/s}. \quad (5.5)$$

Combining equations 5.3 and 5.4, we get:

$$|\vec{B}(\rho, \phi)| = \frac{3}{2} B_r G_{s,3} \left(1 - \frac{\rho_1^3}{\rho_2^3} \right) \frac{\rho^2}{\rho_1^2}. \quad (5.6)$$

The magnetic flux norm is directly proportional to the square of the radial position in the cylinder bore. Field curvature in the bore of the cylinder can be altered by four terms in equation 5.6: B_r , $G_{s,3}$, ρ_1 and ρ_2 .

The magnetic remanence of the magnets employed influences the field curvature of a Halbach cylinder. Sintered rare-earth neodymium iron boron (NdFeB) magnets are currently the strongest permanent magnets available, with a maximum remanence of $B_r \leq 1.4\text{ T}$. To achieve higher flux densities, superconducting magnets are necessary. However, using permanent magnets is often desirable due to their simpler experimental setup.

Figure 5.2a shows the geometry factor for a hexapole cylinder as a function of the number of magnet segments (Eq. 5.5). Creating the six poles requires a multiple of 6 magnet segments ($s = 6, 12, 18, \dots$). Increasing the number of segments leads to higher field gradients, although the returns diminish rapidly. Moreover, a greater number of segments results in more unique magnetisations, thereby increasing the manufacturing cost. Additionally, as the number of segments increases, the individual segments necessarily become smaller. Sintered NdFeB magnets, although capable of being manufactured in non-standard shapes and sizes, are relatively brittle and prone to corrosion in harsh environments due to their iron content¹. Using fewer segments has the practical advantage of reducing the risk of magnet damage during assembly, which is particularly important considering the substantial forces involved. It also reduces safety risks, as high remanence NdFeB magnets with volumes larger than a few cubic centimetres can cause serious injury if mishandled.

Figure 5.2b illustrates the bracketed term in equation 5.6, with the substitution $x = \rho_1/\rho_2$. Since the outer radius of the cylinder is always greater than the inner radius ($\rho_1 < \rho_2$), we have $0 < x < 1$.

¹SmCo alloys serve as a better alternative if a lower magnetic remanence is acceptable.

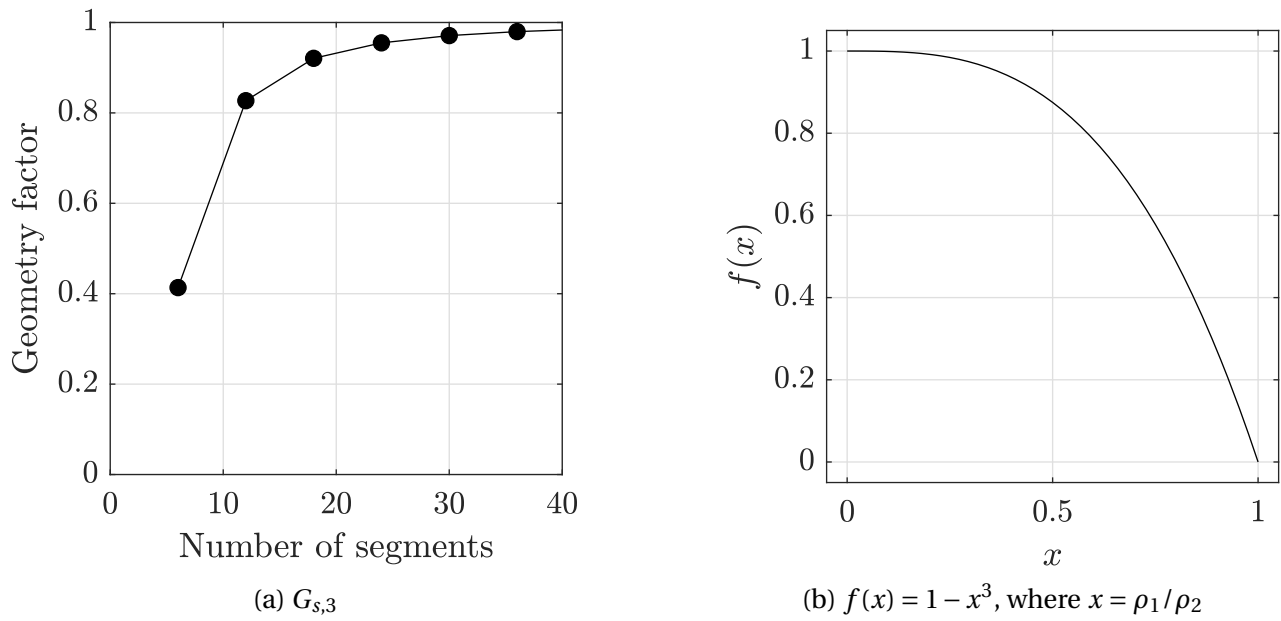


Figure 5.2: Geometry factor for a hexapole cylinder and logarithm of the bracketed term from Eq. 5.6.

For an outer radius twice that of the inner radius, we obtain $1 - x^3 = 0.875$, representing 87.5% of the maximum value. Further increasing the outer radius results in greater field curvature, but the benefits diminish rapidly.

The remaining term to consider in equation 5.6 is $1/\rho_1^2$. Reducing the inner radius of the cylinder enhances the field curvature. However, this also reduces the available space for particle traversal, potentially leading to some particles being obstructed by the cylinder. Striking a balance between field curvature and particle accessibility is further discussed in section 5.2.5.

In sum, achieving high flux density and field curvature in a hexapole magnetic lens requires carefully tuning the magnet properties. The force experienced by a molecule passing through the lens is directly proportional to the magnetic remanence of the permanent magnets. Increasing the number of segments can enhance the flux density, but it comes at the expense of higher costs and an increased risk of damage during assembly. Additionally, it is important to use a small bore and thick magnets, ensuring that the outer radius is significantly greater than the inner radius, ideally $\rho_2 > 2\rho_1$.

5.1.3 A YbF molecule in a hexapole Halbach cylinder

Force on a YbF molecule in a hexapole Halbach cylinder

The force on a neutral YbF molecule in a magnetic field is caused by the Zeeman shift. From section 2.2.1, we find that in the strong field limit, the Zeeman shift varies linearly with magnetic field strength. Fields in excess of 1 T are required to impart enough acceleration on the YbF molecules to create a short, and thus practical, magnetic lens. Therefore, the molecules spend most of their time in the magnetic lens in the strong field regime. The force in the strong field regime is,

$$\vec{F} = -\mu_B \nabla |\vec{B}(\rho, \phi)|. \quad (5.7)$$

Using equation 5.6, for the norm of the magnetic flux density, we get,

$$\vec{F} = -3\mu_B B_r G_{s,3} \left(1 - \frac{\rho_1^3}{\rho_2^3}\right) \frac{\rho}{\rho_1^2} \hat{\rho}. \quad (5.8)$$

Equations of Motion

From the force equation 5.8, we can now proceed with deriving the equations of motion (eoms) for a particle traversing a hexapole lens. Let's start by simplifying the equation with the substitution,

$$\omega^2 = \frac{3\mu_B B_r G_{s,3}}{m} \left(1 - \frac{\rho_1^3}{\rho_2^3}\right), \quad (5.9)$$

where m is the mass of the YbF molecules. Then transforming the force from cylindrical coordinates into Cartesian coordinates (to make comparisons with other Cartesian simulations easier):

$$\vec{F} = -m\omega^2 \rho \hat{\rho} \quad (5.10)$$

$$= -m\omega^2 (x\hat{x} + y\hat{y}). \quad (5.11)$$

Motion in the two transverse axes is uncoupled and can therefore be treated independently. In each axis, molecules will undergo simple harmonic motion (SHM). Applying Newton's second law and solving for the equations of motion (eoms) in the x-axis results in,

$$x(t) = x_0 \cos(\omega t) + \frac{v_{x,0}}{\omega} \sin(\omega t) \quad (5.12)$$

$$v_x(t) = v_{x,0} \cos(\omega t) - x_0 \omega \sin(\omega t) \quad (5.13)$$

where x_0 is initial position of the molecule and $v_{x,0}$ is the initial velocity. The frequency of oscillation, ω , depends on the mass, m , of the molecules and the other parameters previously defined.

Repeating this process for the y-axis results in eoms of identical form and with the same frequency, ω .

5.2 Modelling methods

This section focuses on the modelling methods employed to design our magnetic lens. The primary goal of these simulations was to develop a magnetic lens capable of effectively guiding molecules towards our cooling light source while ensuring an optimal match between the molecular phase space distribution (PSD) and the phase space acceptance of laser cooling. The magnetic lens needed to accommodate molecules with a wide range of longitudinal velocities while also considering the physical constraints imposed by the dimensions of our vacuum chamber.

5.2.1 Finite element analysis of a hexapole magnetic lens

In section 5.1.2, the magnetic flux density of a hexapole lens was defined. Whilst the derivation (see [108, 113]) uses a detailed model, it doesn't account for edge effects or other subtleties of the magnet geometries like filleted edges. These are incredibly difficult to include in an analytical model of the lens. Finite element analysis (FEA) can be used to probe these effects without having to work through laborious and potentially insoluble analytical problems. Typically used for modelling of dynamics like fluid flow and heat transfer, electromagnetic fields are an ideal candidate for FEA. However, it is computationally expensive, requiring a lot of computer memory and taking a long time to compute; both increasing with finite element model (FEM) resolution.

Initial simulations of the magnetic lens were performed using the analytical framework from section 5.1.2. Once a suitable design was identified, COMSOL Multiphysics modeling software was used to perform FEA of the magnetic fields in the lens. The results of this FEM were then used to fine-tune the lens design.

5.2.2 Phase space distributions

Ellipse representation and second order phase space statistics

When modelling a particle beam, using Monte Carlo simulation techniques to emulate the system is common practice. Particles are drawn from their PSD and propagated according to the eoms stipulated by the system. Since particles are being treated independently, non-conservative forces - such as laser cooling - can be modelled. However, this method can be computationally expensive to simulate large numbers of particles. For a system in which only conservative forces are present, one can instead model the PSD itself and propagate it according to the eoms. To model one experimental setup using this method requires only a few calculations, compared to the many thousands of particles that one would normally simulate using a Monte Carlo method.

For a bunch of molecules exiting the aperture of our molecular source, their PSD is approximately

a two-dimensional Gaussian. This can be represented by an ellipse of the general form,

$$0 = Aw^2 + 2Bww' + Cw'^2 + Dw + Ew' + F \quad (5.14)$$

where $w = \{x, y, z\}$ and $w' = \frac{1}{\omega}\{v_x, v_y, v_z\}$ respectively². Parameters A , B and C of the ellipse are related to: the rotation angle, θ , of the ellipse (the angle between the positive w axis and the ellipse's major axis); the semi-major axis, a , and the semi-minor axis, b .

$$A = a^2 \sin^2 \theta + b^2 \cos^2 \theta \quad (5.15a)$$

$$B = (b^2 - a^2) \sin \theta \cos \theta \quad (5.15b)$$

$$C = a^2 \cos^2 \theta + b^2 \sin^2 \theta \quad (5.15c)$$

The enclosed area of the ellipse, known as emittance, ϵ , is constant when only conservative forces are applied to the PSD. If the mean of the phase space distribution, (w_0, w'_0) , is centered on the origin, then $F = -\epsilon$ is the emittance of the PSD. Parameters D and E are defined as,

$$D = -2(A\langle w \rangle + B\langle w' \rangle) \quad (5.16)$$

$$E = -2(B\langle w \rangle + C\langle w' \rangle) \quad (5.17)$$

where $\langle w \rangle$ and $\langle w' \rangle$ are the means of these distributions. Therefore, D and E are zero when the PSD is centered on the origin. This results in the simplified ellipse equation,

²The PSD of a cloud of particles is defined by their momentum and position distributions. However, it is common practice to work in position-position space when working in the modelling framework described here. Appendix A.1 details the transformation between these frames.

$$\epsilon = Aw^2 + 2Bww' + Cw'^2. \quad (5.18)$$

In accelerator physics, where this framework is frequently used to model transverse beam dynamics, the parameters $\{A, B, C\}$ are known as Twiss parameters $\{\gamma, \alpha, \beta\}$ (or Courant – Snyder parameters) [115–121]. To remain consistent with the various accelerator physics literature, let's substitute in these symbols:

$$\epsilon = \gamma w^2 + 2\alpha ww' + \beta w'^2. \quad (5.19)$$

Figure 5.3 shows an example ellipse with key points labelled in terms of Twiss parameters [120, 121]. The Twiss parameters define the shape of the ellipse and are related to the second moments of the PSD:

$$\gamma\epsilon = \langle w'^2 \rangle \quad (5.20)$$

$$\alpha\epsilon = -\langle ww' \rangle \quad (5.21)$$

$$\beta\epsilon = \langle w^2 \rangle \quad (5.22)$$

where the second moments are generalised for a two-dimensional probability phase space distribution, $f_{2D}(w, w')$ [119]. These can be determined as follows:

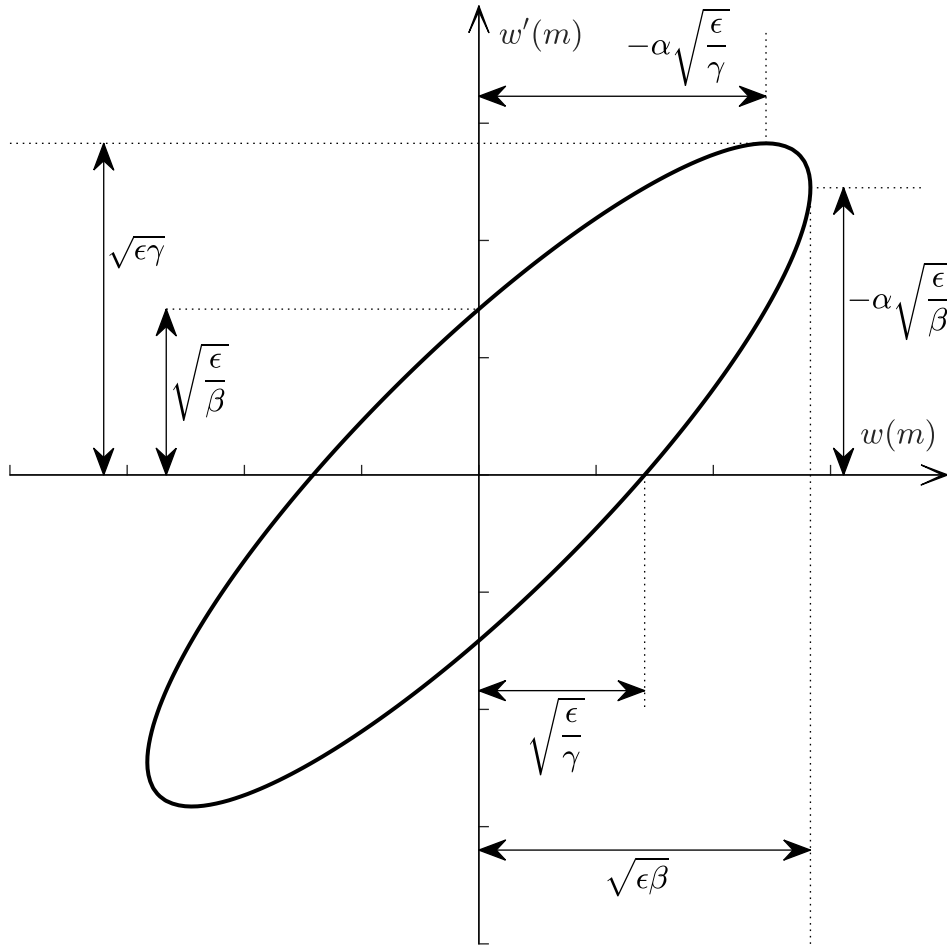


Figure 5.3: Phase space ellipse with Twiss parameter labels.

$$\langle w'^2 \rangle = \frac{\int w'^2 f_{2D}(w, w') dw dw'}{\int f_{2D}(w, w') dw dw'} \quad (5.23)$$

$$\langle ww' \rangle = \frac{\int ww' f_{2D}(w, w') dw dw'}{\int f_{2D}(w, w') dw dw'} \quad (5.24)$$

$$\langle w^2 \rangle = \frac{\int w^2 f_{2D}(w, w') dw dw'}{\int f_{2D}(w, w') dw dw'}. \quad (5.25)$$

For our experiment, a two-dimensional Gaussian distribution is an accurate approximation. The second moments $\langle w^2 \rangle$ and $\langle w'^2 \rangle$ are then the variance of the position and velocity distributions respectively, and $\langle ww' \rangle$ their correlation. All particles within ellipse 5.19 move on their own respective ellipses in phase space which are contained within the ellipse 5.19.

From Wiedemann (2015) [115], we find that this phase space description can be extended to higher

dimensional hyperellipsoids. The equation for an n-dimensional ellipse can be written as follows,

$$\mathbf{u}^T \boldsymbol{\sigma}^{-1} \mathbf{u} = 1 \quad (5.26)$$

where $\mathbf{u}$ is the n-dimensional coordinate vector,

$$\mathbf{u} = \begin{bmatrix} x \\ x' \\ y \\ y' \\ \vdots \\ w_n \\ w'_n \end{bmatrix} \quad (5.27)$$

and $\boldsymbol{\sigma}$ is known as the beam matrix [115]. From comparison with Eq. 5.19, we find the the beam matrix for a two-dimensional phase space ellipse is,

$$\boldsymbol{\sigma} = \begin{bmatrix} \sigma_{11} & \sigma_{12} \\ \sigma_{21} & \sigma_{22} \end{bmatrix} = \begin{bmatrix} \langle w^2 \rangle & \langle w w' \rangle \\ \langle w w' \rangle & \langle w'^2 \rangle \end{bmatrix} = \epsilon \begin{bmatrix} \beta & -\alpha \\ -\alpha & \gamma \end{bmatrix}, \quad (5.28)$$

where we use the identity [115],

$$\beta\gamma - \alpha^2 = 1. \quad (5.29)$$

The beam matrix in Eq. 5.28 is the two-dimensional covariance matrix for motion in one transverse

axis [119]. Its determinant relates the emittance of the ellipse to the second moments:

$$\epsilon = \sqrt{\langle w^2 \rangle \langle w'^2 \rangle - \langle ww' \rangle^2}, \quad (5.30)$$

which, for the two-dimensional Gaussian probability distribution we are using, will be the root mean square (RMS), $\epsilon \rightarrow \epsilon_{\text{RMS}}$.

Considering the 4-dimensional case, $\mathbf{u}^T = (x \ x' \ y \ y')$, we get the beam matrix describing motion in both transverse axes [115, 119]. From Wolski et al. (2020) [119], we find the 4x4 covariance matrix:

$$\sigma = \begin{bmatrix} \langle x^2 \rangle & \langle xx' \rangle & \langle xy \rangle & \langle xy' \rangle \\ \langle xx' \rangle & \langle x'^2 \rangle & \langle x'y \rangle & \langle x'y' \rangle \\ \langle xy \rangle & \langle x'y \rangle & \langle y^2 \rangle & \langle yy' \rangle \\ \langle xx' \rangle & \langle x'y' \rangle & \langle yy' \rangle & \langle y'^2 \rangle \end{bmatrix}. \quad (5.31)$$

This simplifies if the two transverse axes are not coupled to one another [119]. An ideal hexapole Halbach cylinder has no coupling between the two transverse axes, but, when the magnet is segmented into smaller pieces, higher-order poles are introduced, which can cause coupling. However, the strength of these higher-order fields is usually negligible compared to the hexapole field component. Thus our covariance matrix simplifies to,

$$\sigma = \begin{bmatrix} \langle x^2 \rangle & \langle xx' \rangle & 0 & 0 \\ \langle xx' \rangle & \langle x'^2 \rangle & 0 & 0 \\ 0 & 0 & \langle y^2 \rangle & \langle yy' \rangle \\ 0 & 0 & \langle yy' \rangle & \langle y'^2 \rangle \end{bmatrix} \quad (5.32)$$

where we see two 2x2 matrices taking the form given in Eq. 5.28 and no cross-plane elements

$(\langle xy \rangle, \langle xy' \rangle, \langle x'y \rangle \text{ and } \langle x'y' \rangle)$ [119].

5.2.3 YbF phase space distribution

The phase space distribution of our molecules depends on the temperature of the copper cell in which the YbF molecules are being made and on its exit aperture diameter. For a BGS running at temperature T , molecules will be produced with root mean square (RMS) velocity, v_{RMS} in any one direction,

$$v_{\text{RMS}} = \sqrt{\frac{k_B T}{m}} \quad (5.33)$$

where k_B is Boltzmann's constant and m is the mass of a YbF molecule. This is also known as thermal velocity, and its square is the variance of the thermal velocity distribution. Thus we have parameter γ of our ellipse:

$$\gamma = \frac{\langle w'^2 \rangle}{\epsilon_{\text{RMS}}} = \frac{\langle v_w^2 \rangle}{\epsilon_{\text{RMS}} \omega^2} = \frac{v_{\text{RMS}}^2}{\epsilon_{\text{RMS}} \omega^2}. \quad (5.34)$$

Upon exiting the BGS, we assume that the transverse position and velocity distributions are uncorrelated and thus, parameter α of the ellipse is zero. Parameter β relates to the variance in position. The aperture of the copper cell has radius, r_c , of 2 mm. Here, we have assumed that the velocity and position distributions of the molecules are initially uncorrelated ($\theta = 0$ or $\theta = \pi/2$), so we can use equation 5.15c to find the β parameter:

$$\beta = \frac{r_c^2}{\epsilon_{\text{RMS}}}, \quad (5.35)$$

where it is assumed that the molecular position distribution is Gaussian with an rms radius equal to the cell exit aperture radius.

Finally, we can calculate our RMS emittance using equation 5.30:

$$\epsilon_{\text{RMS}} = \sqrt{\langle w^2 \rangle \langle w'^2 \rangle - \langle w w' \rangle^2} \quad (5.36)$$

$$= \sqrt{\langle w^2 \rangle \langle w'^2 \rangle} \quad (5.37)$$

$$= \frac{r_c v_{\text{RMS}}}{\omega} \quad (5.38)$$

Substituting these Twiss parameters in the ellipse equation 5.19 and rearranging results in,

$$r_c^2 = w^2 + \kappa^2 w'^2, \quad (5.39)$$

where,

$$\kappa = \frac{r_c \omega}{v_{\text{RMS}}}. \quad (5.40)$$

Let's examine two examples involving a bunch of YbF molecules, one with a temperature of 5 K and another with a temperature of 15 K for comparison purposes. To illustrate these examples, we need a frequency, ω , for our magnetic lens. For this information, I direct you to the results section 5.3, where $\omega = 1.38 \text{ krad s}^{-1}$. Using this frequency, we can calculate the corresponding κ values for the two examples, which are 189 mrad and 109 mrad respectively. Figure 5.4 displays the contour plots for each ellipse. It is important to note that these contours are valid for both the transverse axes of the beam, x and y , but not for the longitudinal axis, z , which has an average forward velocity exceeding 150 m s^{-1} .

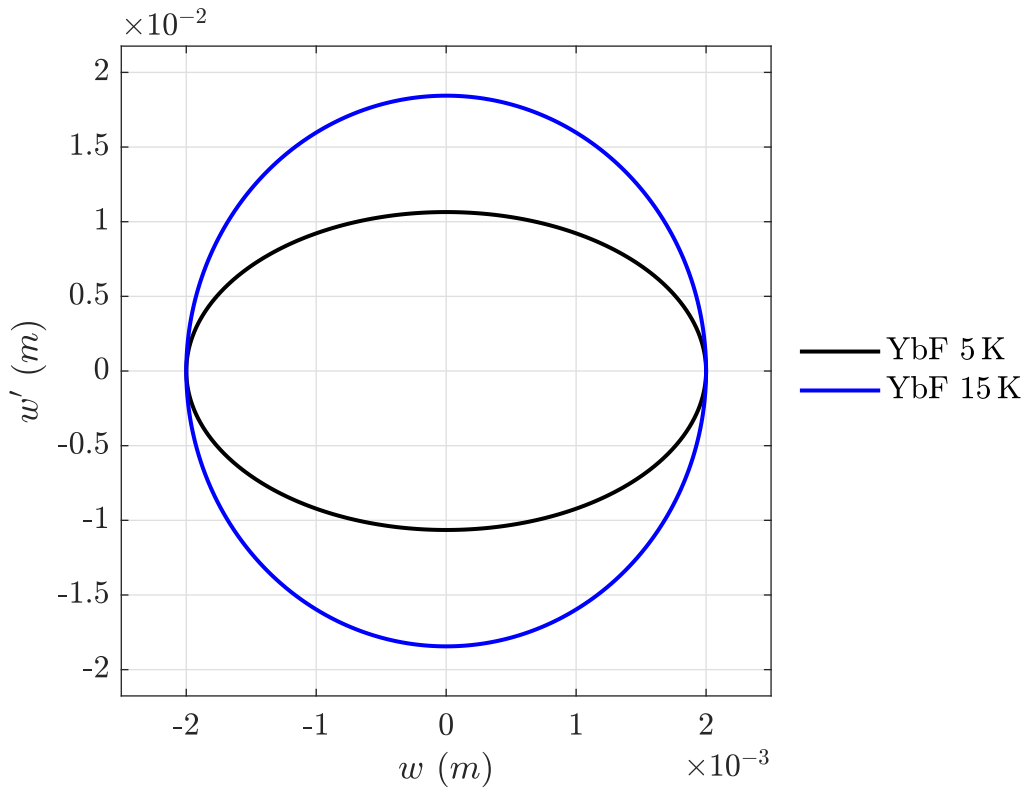


Figure 5.4: Simulated YbF phase space ellipses for different temperatures. The black contour shows a 5 K YbF PSD ellipse, whilst the larger blue contour shows a 15 K YbF PSD ellipse for comparison.

5.2.4 Time evolution of phase space distributions

Free propagation of a molecular beam

Freely propagating molecules will follow these eoms:

$$w(t) = w_0 + w'_0 \omega t \quad (5.41)$$

$$w'(t) = w'_0 \quad (5.42)$$

where $w = \{x, y\}$, w_0 is the initial position, w'_0 is the initial slope in trajectory and t is the propagation time. There is no coupling between the axes of the system, so we can easily determine the second moments using the eoms.

$$\langle w(t)^2 \rangle = \langle (w_0 + w'_0 \omega t)^2 \rangle \quad (5.43)$$

$$= \langle w_0^2 \rangle + 2\omega t \langle w_0 w'_0 \rangle + \omega^2 t^2 \langle w'^2_0 \rangle \quad (5.44)$$

$$\langle w(t) w'(t) \rangle = \langle (w_0 + w'_0 \omega t) w'_0 \rangle \quad (5.45)$$

$$= \langle w_0 w'_0 \rangle + \omega t \langle w'^2_0 \rangle \quad (5.46)$$

$$\langle w'(t)^2 \rangle = \langle w'^2_0 \rangle \quad (5.47)$$

The second moments on the right-hand sides of these equations, $\langle w'^2_0 \rangle$, $\langle w_0 w'_0 \rangle$ and $\langle w^2_0 \rangle$, are related to the initial ellipse parameters γ_0 , α_0 and β_0 , whilst the time evolving second moments on the left of the equations relate to $\gamma(t)$, $\alpha(t)$ and $\beta(t)$ (see equations 5.20 - 5.22). The coefficients of the initial Twiss parameters can be used to form a transformation matrix, $M_{\text{Free}}(t)$, such that,

$$\begin{pmatrix} \gamma(t) \\ \alpha(t) \\ \beta(t) \end{pmatrix} = M_{\text{Free}}(t) \cdot \begin{pmatrix} \gamma_0 \\ \alpha_0 \\ \beta_0 \end{pmatrix}, \quad (5.48)$$

where,

$$M_{\text{Free}}(t) = \begin{pmatrix} 1 & 0 & 0 \\ -\omega t & 1 & 0 \\ \omega^2 t^2 & -2\omega t & 1 \end{pmatrix}. \quad (5.49)$$

Note that the transformation doesn't change the emittance since there is no force on the molecules. More generally, the emittance is conserved unless a non-conservative force is applied.

Figure 5.5 illustrates the time evolution of the ellipse parameters γ , α , and β for an initial ellipse with $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$ and $\epsilon = 1 \text{ m}^2$. The parameter γ remains constant over time since there

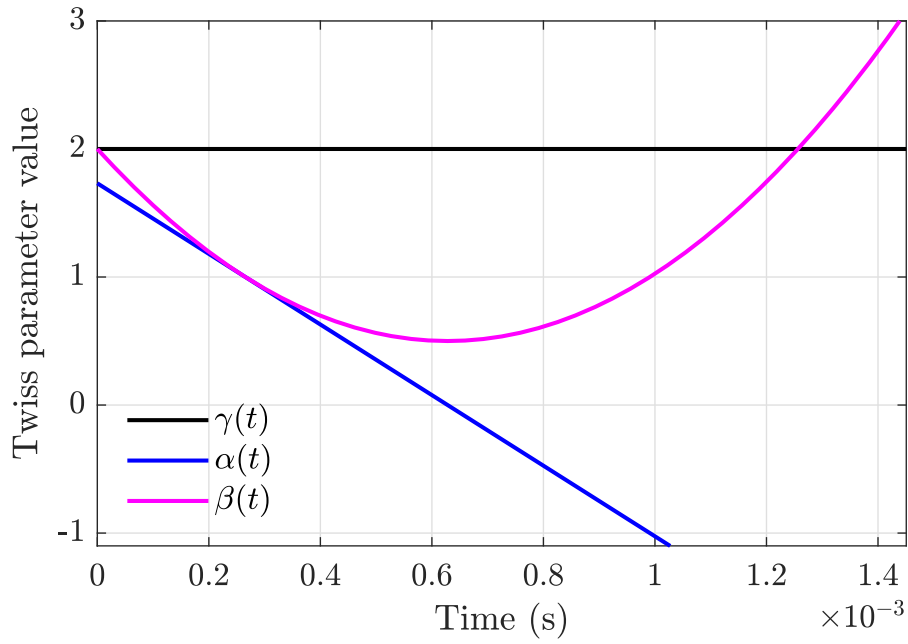


Figure 5.5: Simulated Twiss parameters $\gamma(t)$, $\alpha(t)$ and $\beta(t)$ of a freely propagating PSD with initial parameters $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$, $\epsilon = 1 \text{ m}^2$ and $\omega = 1.38 \text{ krad s}^{-1}$.

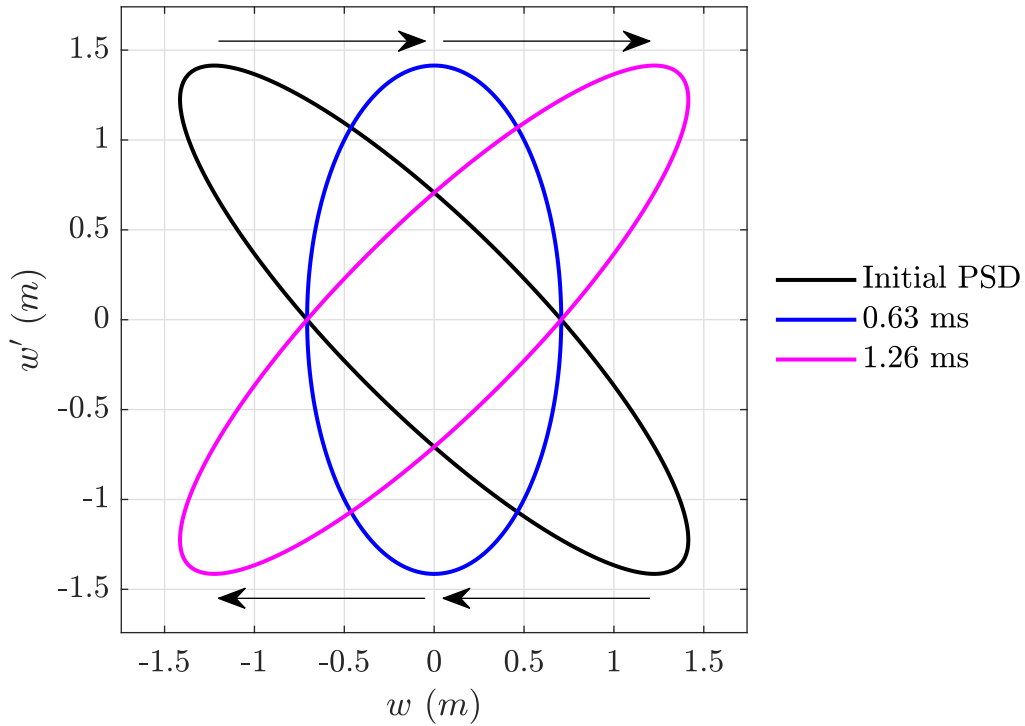


Figure 5.6: An example of a freely propagating PSD with $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$, $\epsilon = 1 \text{ m}^2$ and $\omega = 1.38 \text{ krad s}^{-1}$. The black contour represents the initial PSD, with $\alpha > 0$. As time progresses, the ellipse shears and reaches its focal point at 0.63 ms (blue contour). After this point, the PSD continues to shear, exhibiting continuous divergence. The magenta contour corresponds to the PSD after 1.26 ms of free propagation, where $\alpha(t) = -\alpha_0$.

is no acceleration to alter the velocity. In the case where γ is positive (indicating a non-imaginary ellipse), the ellipse angle decreases over time (shearing occurs), and α decreases monotonically. The molecular beam converges if $\alpha_0 > 0$. At $\alpha = 0$, the molecules reach the focal point and subsequently begin to diverge with $\alpha < 0$. The parameter β varies quadratically with time. When $\alpha_0 > 0$, β decreases until α reaches 0, corresponding to the beam having minimum position width. Afterwards, β increases as the beam diverges. For comparison, figure 5.6 presents phase space ellipses for the same freely evolving beam at three significant moments: the initial ellipse representing a beam of converging particles, the beam at its focal point, and the beam diverging after the focal point.

Propagation of a molecular beam through a hexapole Halbach cylinder

The motion of molecules through a hexapole magnetic lens can be described by the following equations of motion in the transverse axes:

$$w(t) = w_0 \cos(\omega t) + w'_0 \sin(\omega t), \quad (5.50)$$

$$w'(t) = w'_0 \cos(\omega t) - w_0 \sin(\omega t). \quad (5.51)$$

Here, ω represents the frequency of the lens (as given by equation 5.9), and t denotes the time spent propagating through the lens. These equations correspond to the behaviour of a simple harmonic oscillator. The molecules undergo oscillatory motion in the transverse axes of the lens due to the quadratic variation of the magnetic field. In the longitudinal axis (z), where there are no magnetic gradients (assuming an ideal Halbach cylinder), the molecular beam propagates freely without experiencing any forces.

The second moments for these transverse eoms are as follows;

$$\langle w(t)^2 \rangle = \langle (w_0 \cos(\omega t) + w'_0 \sin(\omega t))^2 \rangle \quad (5.52)$$

$$= \langle w_0^2 \rangle \cos^2(\omega t) + \langle w_0 w'_0 \rangle \sin(2\omega t) + \langle w_0'^2 \rangle \sin^2(\omega t) \quad (5.53)$$

$$\langle w(t) w'(t) \rangle = \langle (w_0 \cos(\omega t) + w'_0 \sin(\omega t)) (w'_0 \cos(\omega t) - w_0 \sin(\omega t)) \rangle \quad (5.54)$$

$$= -\langle w_0^2 \rangle \frac{\sin(2\omega t)}{2} + \langle w_0 w'_0 \rangle \cos(2\omega t) + \langle w_0'^2 \rangle \frac{\sin(2\omega t)}{2} \quad (5.55)$$

$$\langle w'(t)^2 \rangle = \langle (w'_0 \cos(\omega t) - w_0 \sin(\omega t))^2 \rangle \quad (5.56)$$

$$= \langle w_0'^2 \rangle \cos^2(\omega t) - \langle w_0 w'_0 \rangle \sin(2\omega t) + \langle w_0^2 \rangle \sin^2(\omega t) \quad (5.57)$$

The time evolution of the Twiss parameters $\gamma(t)$, $\alpha(t)$, and $\beta(t)$ can be determined using these second moments. With the derived expressions, we can now define the transformation matrix, denoted as M_{Lens} , for the magnetic lens. This matrix relates the transformed ellipse parameters to the initial parameters as follows:

$$\begin{pmatrix} \gamma(t) \\ \alpha(t) \\ \beta(t) \end{pmatrix} = M_{\text{Lens}}(\omega, t) \cdot \begin{pmatrix} \gamma_0 \\ \alpha_0 \\ \beta_0 \end{pmatrix}. \quad (5.58)$$

Substituting in the coefficients from the right hand sides of equations 5.52-5.57 and converting the second moments to Twiss parameters using equations 5.20-5.22, we get the transformation matrix³,

$$M_{\text{Lens}}(\omega, t) = \begin{pmatrix} \cos^2(\omega t) & \sin(2\omega t) & \sin^2(\omega t) \\ -\frac{1}{2} \sin(2\omega t) & \cos(2\omega t) & \frac{1}{2} \sin(2\omega t) \\ \sin^2(\omega t) & -\sin(2\omega t) & \cos^2(\omega t) \end{pmatrix} \quad (5.59)$$

³Particular care must be taken when considering the signs of the α and α_0 Twiss parameters.

Figure 5.7 shows the time evolution of the ellipse parameters $\gamma(t)$, $\alpha(t)$, and $\beta(t)$ for a PSD with initial values $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$ and $\epsilon = 1 \text{ m}^2$ propagating through a magnetic lens with a frequency of $\omega = 1.38 \text{ krad s}^{-1}$. As the molecules traverse the lens, they undergo oscillations in the transverse axes. Specifically, $\gamma(t)$ and $\beta(t)$ exhibit harmonic motion, oscillating one-half cycle out of phase. On the other hand, $\alpha(t)$, which corresponds to the angle of the PSD, oscillates out of phase relative to $\gamma(t)$ and $\beta(t)$. Every one-quarter cycle, the major and minor axes of the ellipse align with the coordinate axes, resulting in a periodic pattern.

Figure 5.8 presents an ellipse with initial parameters $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$ and $\epsilon = 1 \text{ m}^2$ propagating through a magnetic lens of frequency $\omega = 1.38 \text{ krad s}^{-1}$. The selected times for the evolution correspond to the red vertical dashed lines in figure 5.7. Initially, the ellipse has $\alpha > 0$ and $\gamma = \beta$. As it rotates clockwise, γ increases while β decreases by an equal amount. When $\alpha = 0$, the major and minor axes of the ellipse align with the (w, w') axes. At this point, γ reaches its maximum value, while β reaches its minimum. The parameters continue to oscillate in this manner throughout the propagation in the magnetic lens.

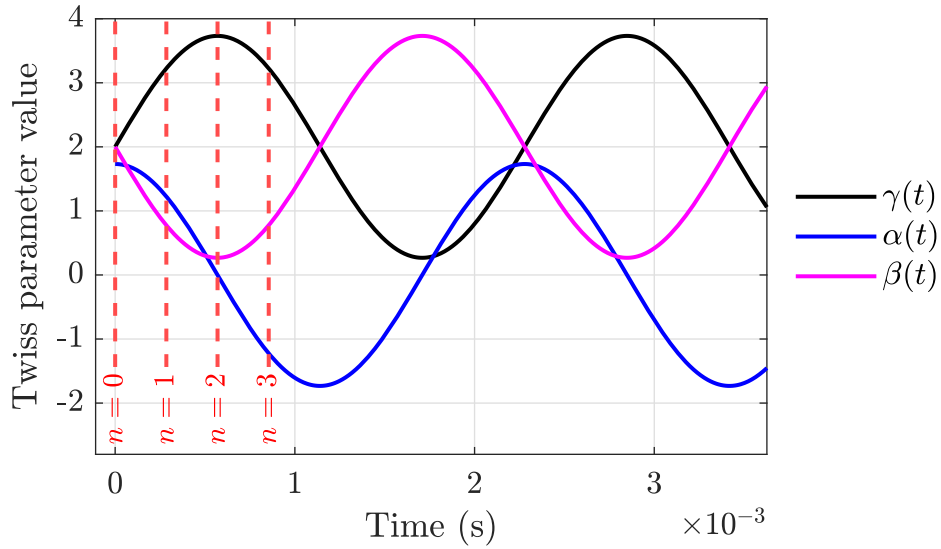


Figure 5.7: Simulated Twiss parameters $\gamma(t)$, $\alpha(t)$ and $\beta(t)$ of a PSD with initial parameters $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$ and $\epsilon = 1 \text{ m}^2$ propagating through a hexapole magnetic lens of frequency $\omega = 1.38 \text{ krad s}^{-1}$. The vertical dashed lines denote the times $t = n\pi/8\omega$ for $n = 0, 1, 2, 3$.

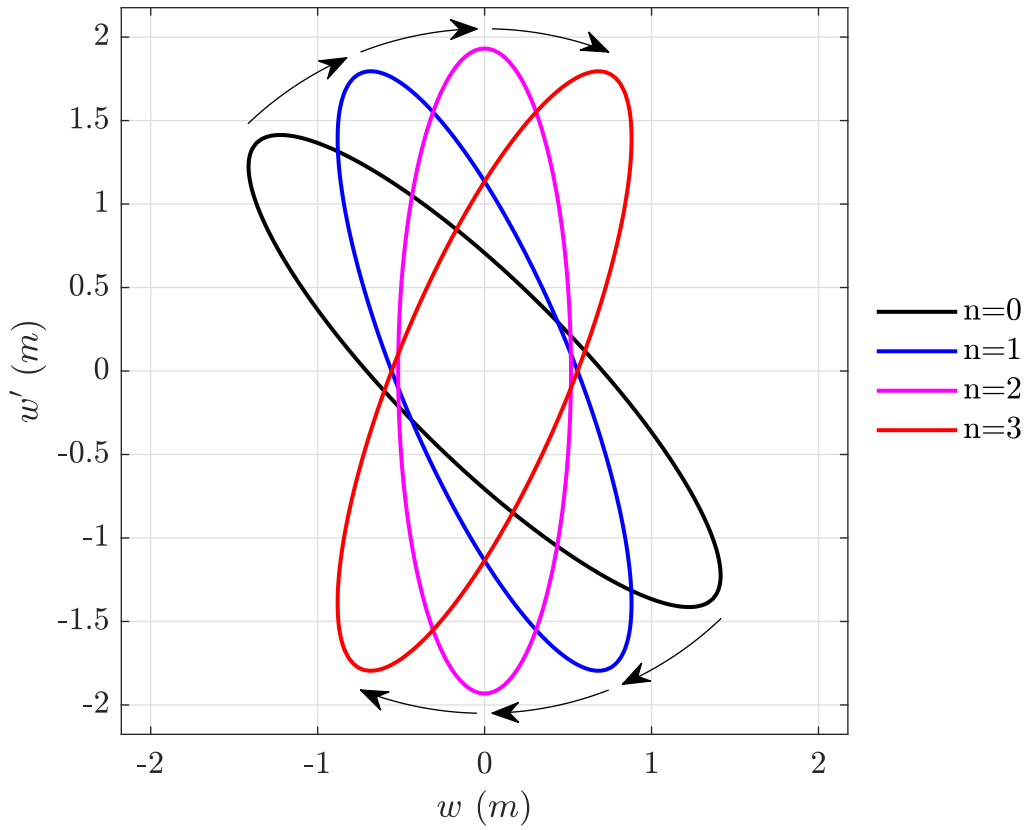


Figure 5.8: Example of a PSD with $(\gamma_0, \alpha_0, \beta_0) = (2, \sqrt{3}, 2)$ and $\epsilon = 1 \text{ m}^2$ propagating through a magnetic lens of frequency $\omega = 1.38 \text{ krad s}^{-1}$. The black contour shows the initial PSD, with $\alpha > 0$. The other contours show the PSD after evolving for times $t = n\pi/8\omega$, where $n = 0, 1, 2, 3$. It rotates as it evolves in time.

5.2.5 Phase space acceptance

The experiment encompasses multiple elements that impose restrictions on the phase space distribution of the molecular beam. The small aperture of the magnetic lens restricts the passage of molecules with large transverse positions. Likewise, laser cooling targets molecules within a limited range of transverse velocities, necessitating their traversal through the cooling light. Each experiment element has its own phase space acceptance, delineating the permissible region of phase space for optimal performance. Failure to match the molecular phase space distribution with the acceptance criteria of a particular element can impede the desired effect of that element.

Magnetic lens phase space acceptance

In section 5.2.4, we explored the evolution of a molecular phase space distribution as it propagates through a magnetic lens. The derived equations of motion (equations 5.50 and 5.51) behave like a harmonic oscillator centred around the axis of the cylinder. However, it is crucial to consider the limitations imposed by the lens's physical structure. Molecules oscillating with excessive amplitudes can collide with the inner surface of the cylinder, resulting in their loss from the experiment⁴. Additionally, molecules entering the lens with transverse positions beyond the radius of the cylinder bore will impact the walls and be lost. These restrictions on the transverse position act as boundary conditions for the molecule's propagation through the lens. By incorporating these boundary conditions with the equations of motion, we can determine the phase space acceptance of the magnetic lens.

At the extremity of its oscillation, the velocity of a molecule vanishes. If we solve $w(t) = 0$ for t , we get the (periodic) times, t_{peak} , at which the molecule will reach the peaks/troughs of its motion:

$$t_{\text{peak}} = \frac{1}{\omega} \left(\arctan \left(\frac{w'_0}{w_0} \right) + 2\pi n \right) \quad (5.60)$$

where $n = 0, 1, 2, \dots$ is zero or a positive integer. Substituting this into the position eom gives us the peak transverse position, w_{peak} , of the molecule:

$$w_{\text{peak}} = \sqrt{w_0'^2 + w_0^2}. \quad (5.61)$$

The physical restriction imposed by the bore radius, r_b , of the cylinder limits the peak transverse position of the molecule: $w_{\text{peak}} \leq r_b$. Molecules with a transverse position exceeding this limit, either before or inside the cylinder, will be lost from the experiment. By considering the extreme

⁴YbF molecules are highly reactive, and any contact with experimental surfaces will almost certainly lead to chemical reactions, resulting in their loss from the experiment.

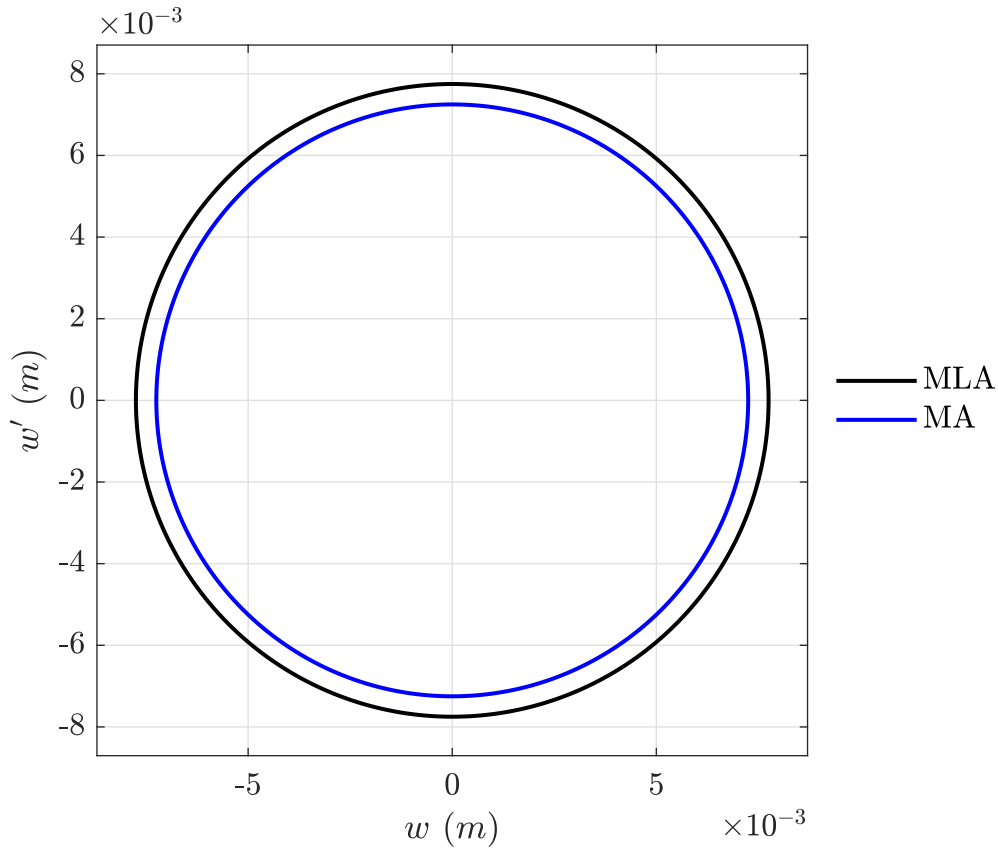


Figure 5.9: The black contour shows the phase space acceptance for a 1.38 krad s^{-1} magnetic lens with bore radius 7.75 mm. The smaller blue contour shows the phase space acceptance for a mount with bore radius 7.25 mm.

case of $w_{\text{peak}} = r_b$ and squaring both sides of equation 5.61, we obtain:

$$r_b^2 = w_0'^2 + w_0^2. \quad (5.62)$$

Equation 5.62 represents a circle and defines the phase space acceptance for neutral molecules propagating through a hexapole magnetic lens. I will refer to this as the magnetic lens acceptance (MLA). By comparing the coefficients of the ellipse parameters with the equation for an ellipse (equation 5.19), we can identify the Twiss parameters as $(\gamma, \alpha, \beta) = (1, 0, 1)$, and the emittance as $\epsilon = r_b^2$.

It is important to note that the velocity capture range is independent of the bore radius of the lens. Increasing the bore radius, r_b , expands the position capture range while leaving the velocity

capture range unchanged. This can be understood by considering the behaviour of a particle travelling through the lens. Increasing the bore radius reduces the magnetic flux density, resulting in a lower lens frequency. Consequently, the particle experiences a reduced force but over a larger distance while the overall work done remains constant. Therefore, the velocity capture range remains invariant with respect to the bore radius.

However, the acceptance of the lens is further constrained by the mount that the magnets are housed in; I refer to this as the mount acceptance (MA). Equation 5.62 describes the phase space acceptance of the mount, except the bore radius of the mount is smaller. Figure 5.9 illustrates an example of the phase space acceptance for a magnetic lens with a frequency of 1.38 krad s^{-1} and a bore radius of 7.75 mm, along with the corresponding mount acceptance for a mount bore radius of 7.25 mm.

Laser cooling phase space acceptance

The magnetic lens is designed to rotate the molecular phase space distribution to overlap the laser cooling phase space acceptance. To model this alignment, we need to know both phase space distributions: the molecular phase space distribution discussed in section 5.2.3, and the laser cooling phase space acceptance, which is outlined here.

Measuring the transverse phase space distribution of an atomic or molecular beam is possible [118, 119]. In principle, one could infer the laser cooling phase space acceptance for our experiment by comparing measurements of the molecular phase space distributions before and after laser cooling. However, accurately measuring the four-dimensional (x, v_x, y, v_y) transverse phase space distribution is challenging [118, 119]. Therefore, we rely on laser cooling simulations to guide the design of our magnetic lens.

Jongseok Lim has conducted trajectory simulations of transverse sub-Doppler laser cooling for *YbF* molecules [1]. These simulations incorporate the non-conservative forces required for laser cooling, which are not accounted for in the modelling methods presented in this work. From Lim's simulations, we can determine the laser cooling acceptance (LCA).

In the trajectory simulations, a broad and uniform molecular phase space distribution is propagated through the cooling light. Molecules that undergo laser cooling experience a reduction in their transverse velocities. We consider molecules with transverse velocities $v_x, v_y < 0.1 \text{ ms}^{-1}$ to be ultracold. By tracing the trajectories of these ultracold molecules back to the entrance of the laser cooling region, we can define the laser cooling phase space acceptance. Note that Lim's simulations were not performed in the w, w' phase space used in this work but instead in position-velocity phase space.

Figure 5.10a presents the raw laser cooling acceptance data obtained from Lim's simulation. It is important to note that the transverse velocity criterion used to capture molecules in the LCA also includes molecules with low transverse initial velocities but outside the capture range of the cooling lasers. To obtain an accurate representation of the phase space acceptance of the cooling forces, these outliers were removed.

To determine the boundary of the LCA data points, a convex hull approach was employed. The convex hull is a curve encompassing the data points with the minimum perimeter. It can be visualised as a rubber band stretched around the data points, tightly hugging them. Figure 5.10b displays the LCA data points along with their corresponding convex hull, which represents the bounds of the phase space acceptance.

Next, an ellipse was fitted to the boundary points of the convex hull. This ellipse provides a concise representation of the acceptance region. Figure 5.10c shows the ellipse fit to the convex hull boundary.

To bring the results of Lim's trajectory simulation into the modelling regime used in this work, the ellipse was transformed into the $\{w, w'\}$ phase space. This transformation was accomplished using a lens frequency of $\omega = 1.38 \text{ krad s}^{-1}$ (see appendix A.1 for details of the transformation). The resulting ellipse in the $\{w, w'\}$ phase space is displayed in Figure 5.10d. It possesses Twiss parameters $\{\gamma, \alpha, \beta\} = \{5.3, 3.6, 130\} \times 10^{-7}$ and an emittance $\epsilon = 2.1 \times 10^{-11} \text{ m}^2$. This transformed ellipse represents the laser cooling phase space acceptance within the context of the current modelling approach.

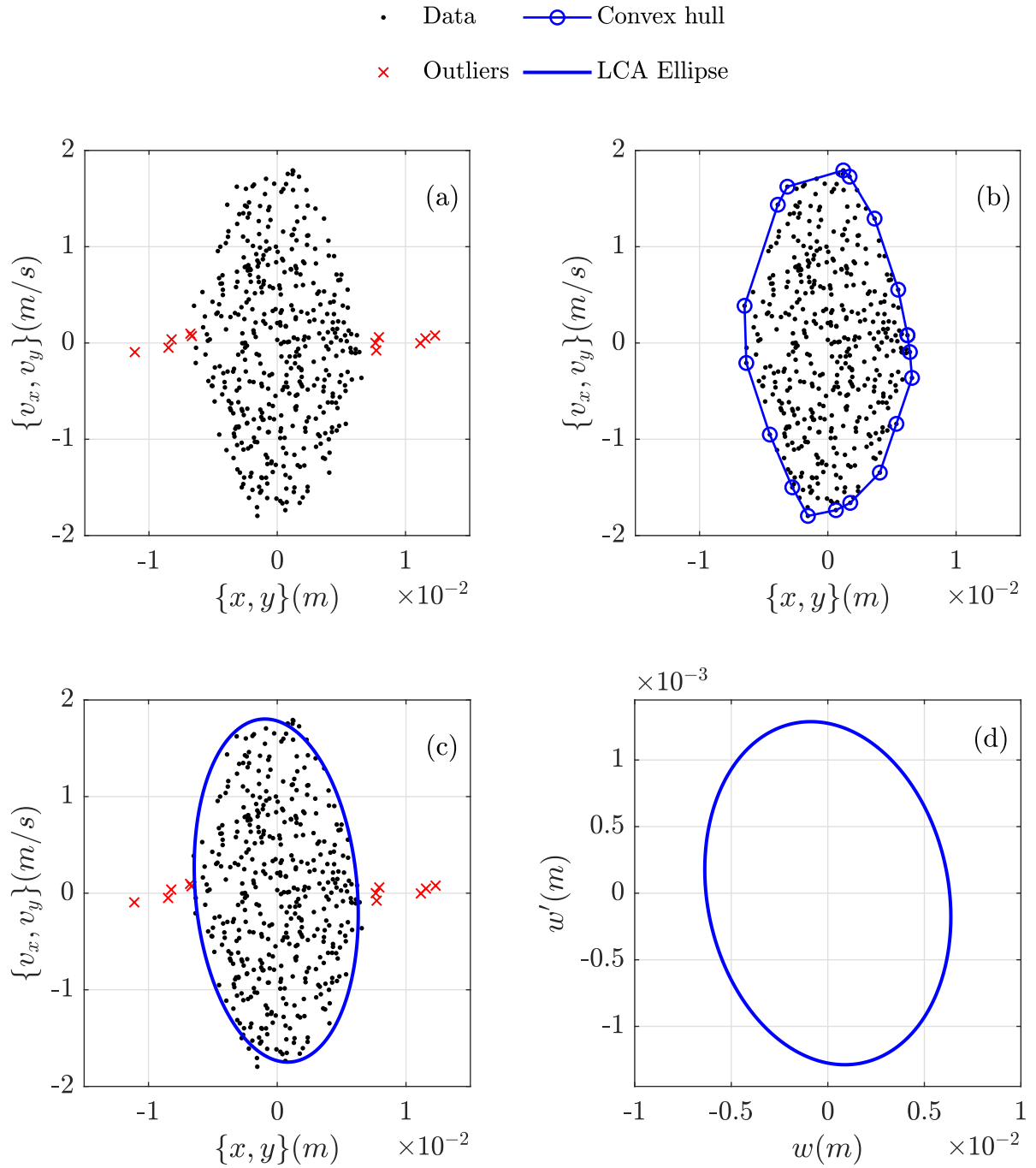


Figure 5.10: Analysis of simulated laser cooling phase space acceptance. a) Raw LCA data. Red crosses mark the outliers which do not travel through the cooling light but do meet the criterion $\{v_x, v_y\} < 0.1 \text{ ms}^{-1}$. b) Raw LCA data with the outliers removed. The blue circles show the points which form the convex hull - connected by a blue line to show the convex hull boundary. c) Raw LCA data with the outliers included for comparison. The blue ellipse is the result of a fit to the convex hull boundary points. d) The ellipse from plot (c) after being transformed to the (w, w') phase space using lens frequency $\omega = 1.38 \text{ krad s}^{-1}$. The resulting ellipse has Twiss parameters $\{\gamma, \alpha, \beta\} = \{5.3, 3.6, 130\} \times 10^{-7}$ and emittance $\epsilon = 2.1 \times 10^{-11} \text{ m}^2$.

5.2.6 Simulating the magnetic lens

The objective of this simulation was to determine the optimal design parameters for our magnetic lens, considering the limitations imposed by our experimental setup, the phase space distribution of our molecular beam, and the phase space acceptance of our cooling light. Since the laser cooling phase space acceptance is relatively small compared to the molecular phase space distribution, there is no single set of design parameters that can be considered universally optimal for the lens. Therefore, the simulation aimed to explore the broad multi-dimensional space to identify a suitable range of parameters.

In this section, I outline the model that I sought to optimise, define the condition that must be satisfied for the lens to be considered "optimised", and discuss the constraints that the model must adhere to. By carefully considering these factors, one can determine the most favourable design parameters for the magnetic lens, ensuring effective performance within the limitations of our experimental system.

Modelling the molecular beam, magnetic lens and laser cooling

The molecular source produces a beam colder than 5 K. Its phase space distribution, outlined in section 5.2.3, propagates freely until it reaches the magnetic lens. The molecules take time, t_1 , to travel this distance. They then spend time, t_2 , propagating through the lens, which has frequency, ω . Upon exiting the lens, the molecules travel freely again, taking time, t_3 , to reach the cooling light. The transformation matrices outlined in section 5.2.4 are applied sequentially to find the transformation for this whole process:

$$\begin{pmatrix} \gamma \\ \alpha \\ \beta \end{pmatrix} = M_{\text{Free}}(t_3) \cdot M_{\text{Lens}}(\omega, t_2) \cdot M_{\text{Free}}(t_1) \cdot \begin{pmatrix} \gamma_0 \\ \alpha_0 \\ \beta_0 \end{pmatrix}, \quad (5.63)$$

where $(\gamma_0, \alpha_0, \beta_0)$ are the Twiss parameters of our initial YbF molecular phase space distribution.

Figure 5.11 depicts a (black) phase space ellipse representing a bunch of YbF molecules with temperature 5 K, travelling with longitudinal velocity 180 ms^{-1} , propagating through the apparatus to the laser cooling region. They propagate freely to the entrance of a lens 10 cm away from the molecular source. Upon reaching the lens, some of the molecules will impact it and be lost from the experiment. Only molecules which reside within the region of phase space bounded by the magnetic lens mount acceptance (blue ellipse⁵) will successfully be guided by the lens. The shaded region depicts the overlap of the ellipses; only these molecules will exit the lens. To follow these molecules to the laser cooling region, we must propagate the mount acceptance in the same way as the molecules - using the transformation $M_{\text{Free}}(t_3) \cdot M_{\text{Lens}}(\omega, t_2)$. Through the lens, the mount acceptance will rotate, and since it is a circle, it will not change. However, at the end of the lens, the mount acceptance will be propagated freely and will shear. Throughout these periods of propagation, the overlap region continues to represent the set of molecules which have traversed the lens. Once the molecular and mount acceptance ellipses have been propagated to the cooling lasers, we must consider their overlap with the laser cooling acceptance ellipse - it is the molecules in this phase space region which will be successfully laser cooled.

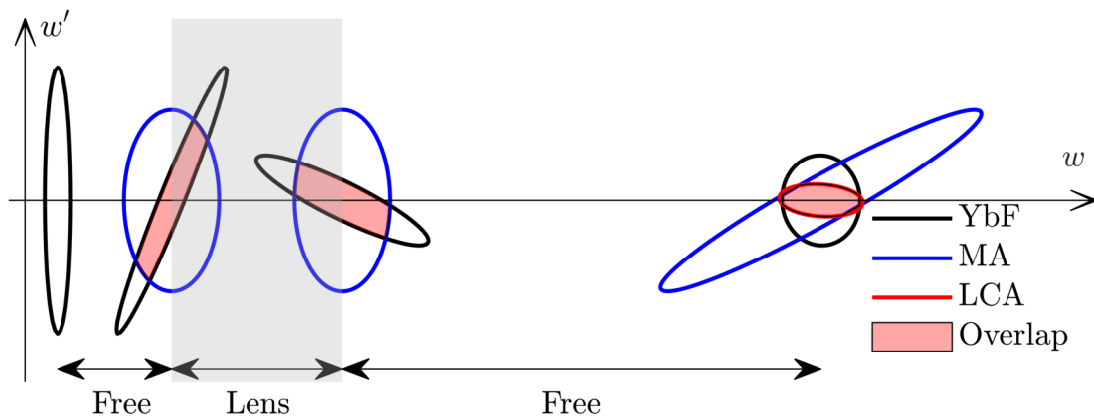


Figure 5.11: Illustration of the magnetic lens simulation. The molecular phase space ellipse (black) propagates freely, then through the lens and then freely again before reaching the laser cooling region. The magnetic lens mount phase space acceptance (blue) propagates through the lens and then freely to the cooling lasers. At the laser cooling region entrance, the laser cooling phase space acceptance is shown (red ellipse). Red-shaded regions show the respective overlaps of the sets of ellipses.

⁵The mount acceptance is a circle, but the scale of the axes in figure 5.11 make it appear elliptical.

The goal of this work was to maximise the number of laser-cooled molecules. Since the emittance of the molecular phase space ellipse cannot be changed by the forces present in this simulation, the maximal number of laser-cooled molecules is achieved when the molecular and mount acceptance ellipses overlap the laser cooling acceptance ellipse completely. In figure 5.11, we see that almost the entirety of the laser cooling acceptance ellipse is shaded. Therefore, this hypothetical lens design would nearly maximise the number of laser-cooled molecules.

It is the overlap of all three phase space ellipses that tells us which molecules make it through the lens before being successfully laser cooled. As the laser cooling phase space acceptance is smaller than the molecular and mount acceptance ellipses, it defines the upper limit for their overlap. Therefore, I define the following figure of merit for the simulation:

$$O_{\text{LCA}} = 100 \times \frac{\text{Overlap Area(YbF, MA, LCA)}}{\text{Area(LCA)}} \quad (5.64)$$

which is the percentage of the laser cooling acceptance which is overlapped with the molecular and mount acceptance ellipses.

Optimization constraints

The wide parameter space must be constrained to make this a tractable problem. Table 5.1 outlines these parameters alongside their respective symbols and values/constraints. Here I discuss the various constraints that were used to simplify the optimisation problem.

Section 5.1.2 outlines the rationale behind some of the constraints applied to the model. Firstly, the outer radius of the magnet segments should be larger than twice the inner radius if we are to maximise the magnetic flux density in the borehole. An outer radius, $\rho_2 \approx 37$ mm, was chosen. This is as wide as the vacuum chamber allows (accounting for the mounting structure). A 12-segment design, $s = 12$, was favoured over one with more segments, sacrificing a little magnetic flux for affordability and structural integrity. Maximal magnetic remanence was chosen for these

Parameter	Symbol	Value/Constraint
Magnet segment outer radius	ρ_2	37 mm
Magnet segment inner radius	ρ_1	$0 \text{ mm} \leq \rho_1 \ll \rho_2$
Lens length	l_{lens}	$10 \text{ cm} \leq l_{\text{lens}} + z_{\text{lens}} \leq 30 \text{ cm}$
Segment number	s	12
Magnetic remanence	B_r	1.44 T
Lens mount inner radius	$\rho_{1,\text{mount}}$	$\rho_{1,\text{mount}} = \rho_1 - 0.5 \text{ mm}$
Source - LCA separation	z_{LCA}	67 cm
Source - Lens separation	z_{lens}	$10 \text{ cm} \leq z_{\text{lens}} \leq 30 \text{ cm} - l_{\text{lens}}$
Beam longitudinal velocity	v_z	$150\text{-}200 \text{ m s}^{-1}$

Table 5.1: Values and/or constraints for the parameters in this simulation. Longitudinal distances are relative to the molecular source.

simulations to maximise the magnetic flux ($B_r = 1.44 \text{ T}$ based on quotes from a magnet supplier).

The current separation of the molecular source and the cooling lasers is 57 cm. However, the cryogenic radiation shields and vacuum chamber components restrict the range of positions where the lens can be mounted. To provide space for the lens, a 10 cm nipple will be added to the vacuum chamber. This will extend the separation of the molecular source and cooling lasers to $z_{\text{LCA}} = 67 \text{ cm}$. Considering the ellipse transformation equation 5.63, we have a total propagation time, $t = t_1 + t_2 + t_3$. So for constant v_z , we have the constraint $v_z t = z_{\text{LCA}} = 67 \text{ cm}$. This results in: $t_1 = z_{\text{lens}}/v_z$, $t_2 = l_{\text{lens}}/v_z$ and $t_3 = (z_{\text{LCA}} - z_{\text{lens}} - l_{\text{lens}})/v_z$. Since z_{LCA} is constant, the three variables z_{lens} , l_{lens} and v_z fully define the times t_1 , t_2 and t_3 .

Finally, we consider the bore radius of the lens, ρ_1 (which in turn constrains the mount bore radius). Increasing ρ_1 will increase the position capture range of the lens and decrease its frequency, ω - increasing the optimal lens length, l_{lens} . Whilst the magnetic flux density will decrease, the transverse propagation distance will increase, and so the work done on a molecule remains unchanged. This means that the velocity capture range of the lens is invariant to changes in ρ_1 , but the position capture range is not.

5.3 Simulation results

The simulation results are presented in two sections: analytical and finite element models. The analytical model is based on the framework developed by Halbach for simulating the magnetic lens, as described in section 5.1.2 [108]. Initially, the analytical model was employed to explore various parameter combinations and identify a promising lens design.

After selecting a suitable lens design based on the analytical model, finite element modelling was conducted to validate the analytical model's results. This involved performing detailed simulations using finite element analysis techniques, as outlined in section 5.2.1. By comparing the results obtained from the analytical model with those from the finite element model, one can ensure the reliability and robustness of the lens design.

5.3.1 Analytical model results

In this section, the results of the analytical modelling are presented. The lens dimensions and position were varied within the bounds of our experimental setup - maximising O_{LCA} for a broad range of beam longitudinal velocities, lens positions and lens lengths. Due to the nuanced constraints and analysis methods, regression methods were deemed impractical.

A few values of ρ_1 were trialled until a lens of reasonable length was found with a broad range of optimal positions and meeting the constraint $10 \text{ cm} \leq l_{\text{lens}} + z_{\text{lens}} \leq 30 \text{ cm}$. A value of $\rho_1 = 7.75 \text{ mm}$ was found to provide desirable lens characteristics - nearly $O_{LCA} = 100\%$ for a broad range of lens positions and lens lengths. The resulting lens frequency then is $\omega = 1.38 \text{ krad s}^{-1}$ and the mount bore radius $\rho_1 - 0.5 \text{ mm} = 7.25 \text{ mm}$.

Figure 5.12 shows O_{LCA} as a function of lens length and lens position (relative to the molecular source) for molecules with a forward velocity $v_z = 180 \text{ ms}^{-1}$. For a given lens length, there is a broad range of optimal positions. Choosing a lens length of 15 cm is convenient as this is easily divisible into 5 cm long segments, a limit set by the manufacturer. This length also offers a broad range of positions for the lens whilst meeting the constraint for z_{lens} listed in table 5.1.

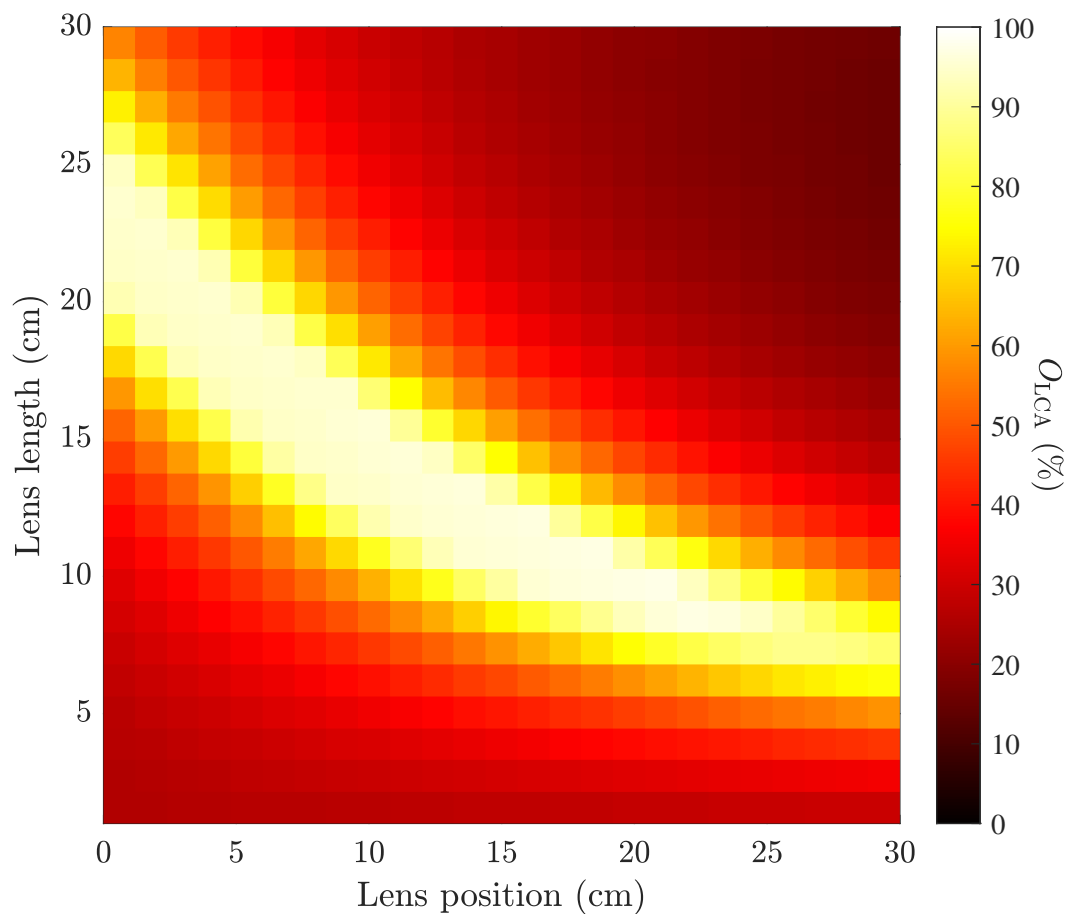


Figure 5.12: Simulation of O_{LCA} as a function of lens position and lens length, for a molecular beam of forward velocity 180 ms^{-1} .

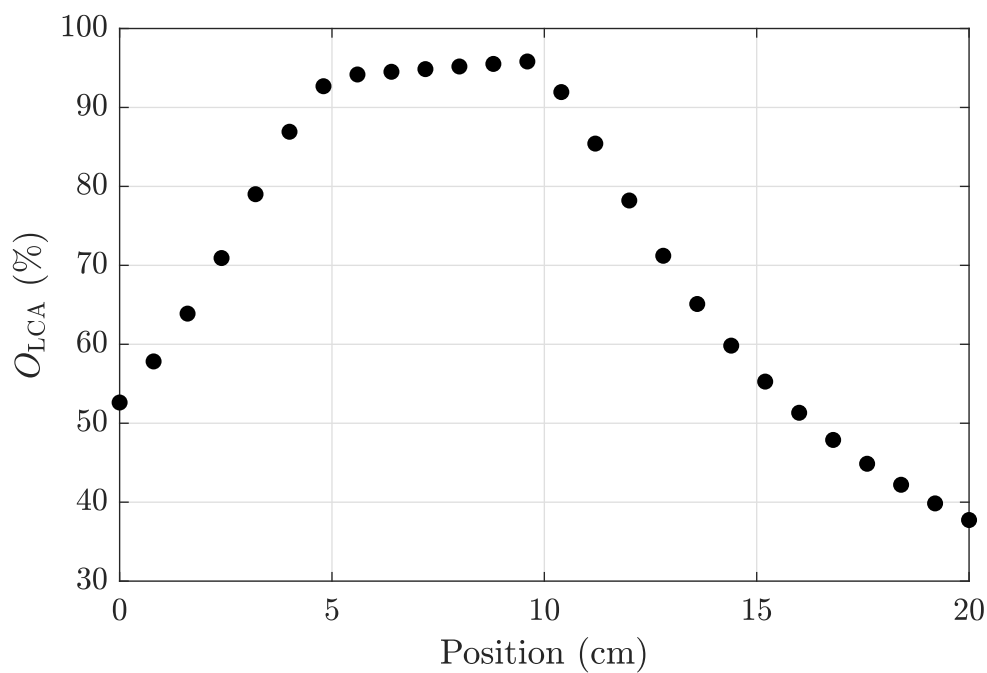


Figure 5.13: Simulation of O_{LCA} as a function of lens position, for a lens of length 15 cm and a molecular beam of forward velocity 180 ms^{-1} .

Figure 5.13 shows a cut of figure 5.12 for a lens length of 15 cm. The overlap area of the phase space ellipses relative to the LCA ellipse area is $>90\%$ over the position range 4.4-10.6 cm. This means that the efficiency of the lens for molecules of this forward velocity is relatively insensitive to the position of the lens - making installation easier.

Figure 5.14 shows how O_{LCA} varies with l_{lens} and v_z , for $z_{\text{lens}} = 8$ cm. For some l_{lens} values, molecules with a broad range of longitudinal velocities can be efficiently guided to the LCA. The choice of $l_{\text{lens}} = 15$ cm here results in $O_{\text{LCA}} >90\%$ over the v_z range 166-213 m s^{-1} .

The plots in figure 5.15 demonstrate the dependency of O_{LCA} on the beam's forward velocity and the longitudinal position of the lens. For a lens of length 15 cm, the lens position was varied over the range 9 – 13 cm and O_{LCA} plotted as a function of v_z . We see that as the lens is moved towards the molecular source, molecules with lower v_z are more efficiently guided to the LCA, with only a small reduction in O_{LCA} . This is useful as a means to fine-tune the experimental setup - the lens can be moved to better guide molecules with forward velocities suitable for laser cooling. Furthermore, the plots demonstrate that the O_{LCA} remains high for a broad range of forward velocities and lens positions.

5.3.2 Finite element model results

The results of the analytical modelling in section 5.3.1 provided a starting point for FEA. COMSOL Multiphysics was chosen for performing this analysis of the magnetic flux density. CAD drawings of the magnet segments were drawn with the dimensions provided in table 5.2. Note that a fillet has been applied to the segments (on the longitudinal edges). Unlike the analytical model, the FEM can easily account for this manufacturing requirement. Twelve 5 cm long segments were assembled with the stainless steel mounts in CAD software and imported to COMSOL for the FEM.

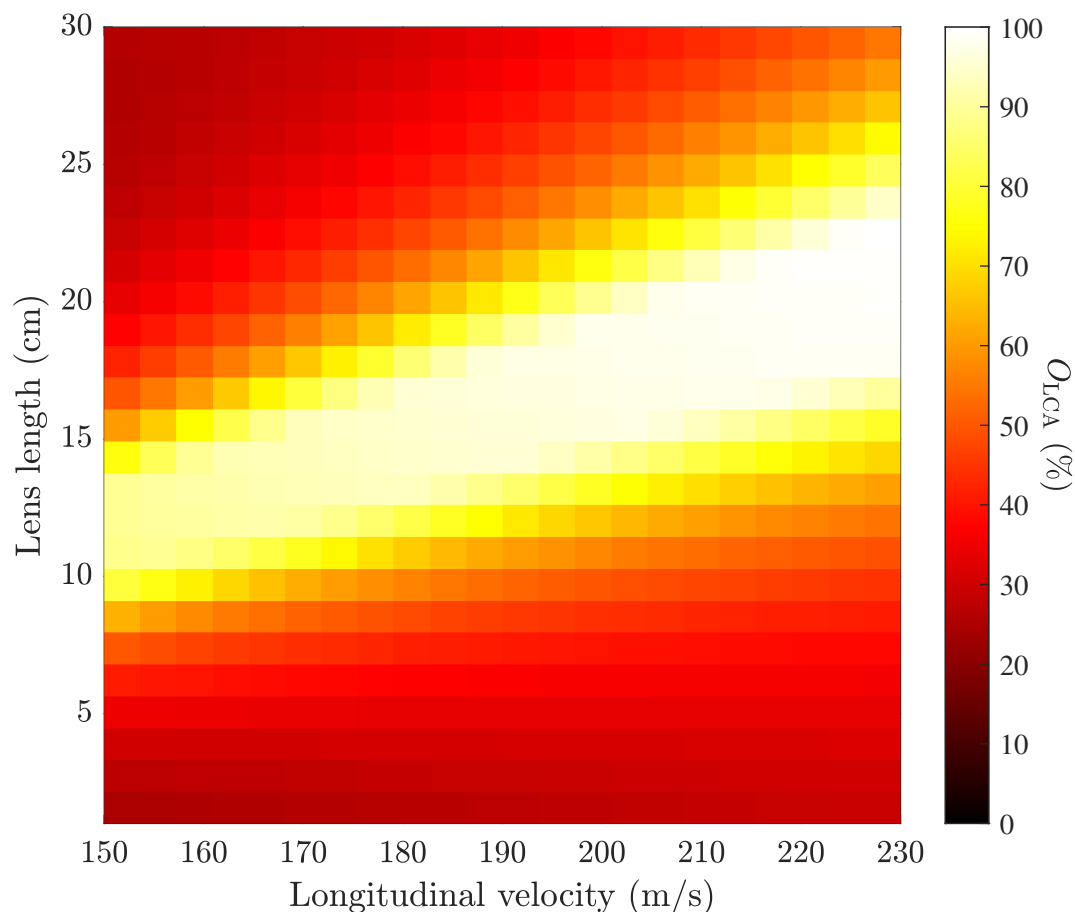


Figure 5.14: Simulation of O_{LCA} as a function of molecular beam forward velocity and lens length, for a lens positioned 8 cm from the molecular source.

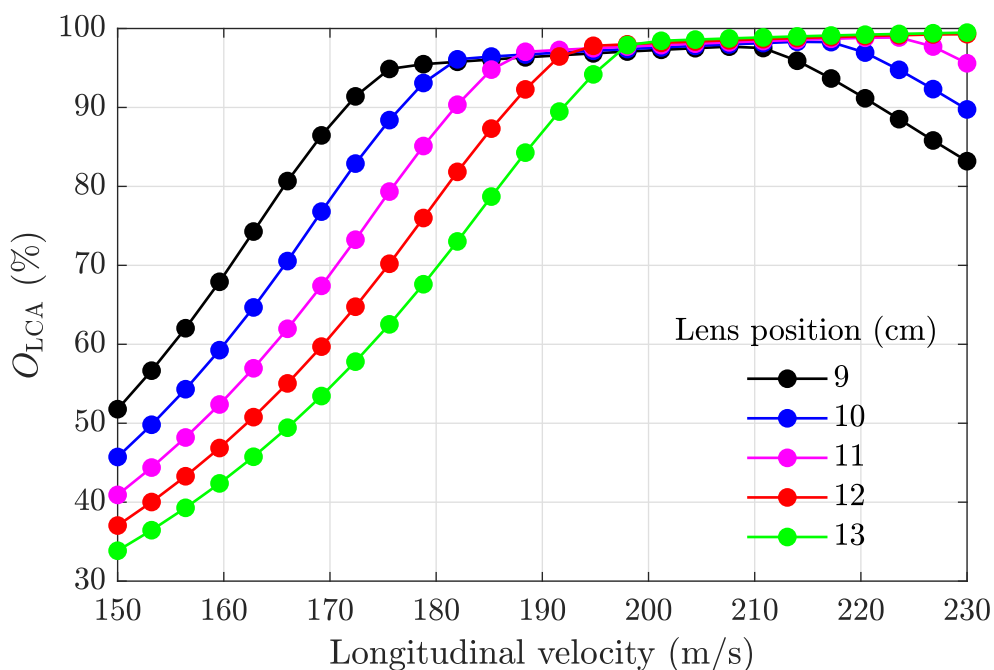


Figure 5.15: Simulation of O_{LCA} as a function of the molecular beams forward velocity, for a lens of length 15 cm, positioned 9 – 13 cm from the molecular source.

Parameter	Symbol	Value
Magnet segment outer radius	r_o	37.25 mm
Magnet segment inner radius	r_i	7.75 mm
Magnet segment length	l_s	5 cm
Fillet radius	r_f	0.5 mm

Table 5.2: CAD drawing parameters for FEM. See section 5.4 for the final design.

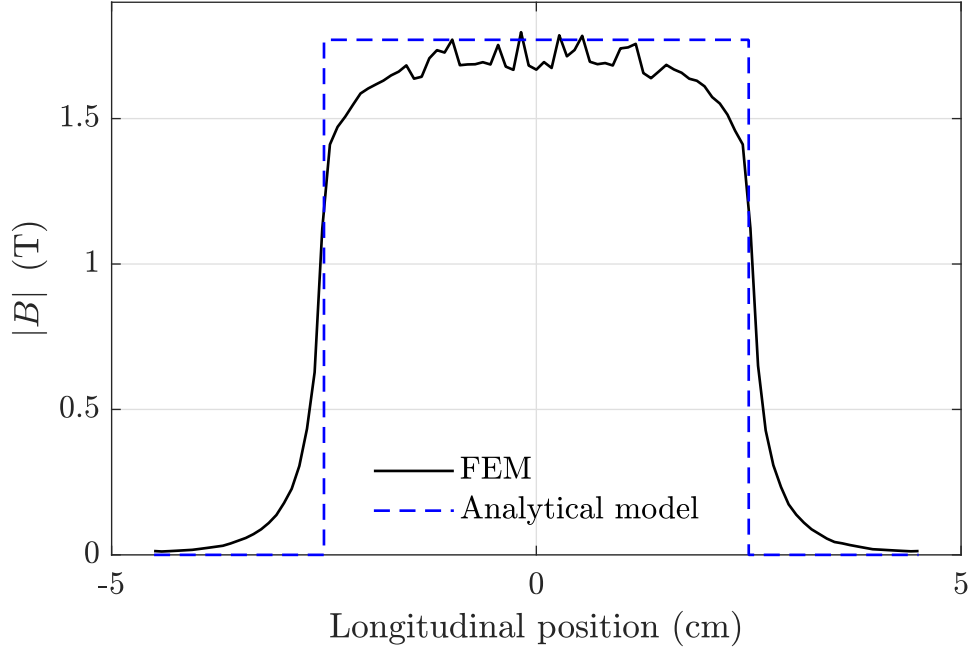


Figure 5.16: Simulation of the magnetic flux density norm as a function of longitudinal position for $(x, y) = (7.75, 0)$ mm (the edge of the borehole). The solid black line shows the finite element model result, while the dashed blue line uses the analytical model for $|B|$. The longitudinal position is centred in the middle of the 5 cm long section of the lens.

Figure 5.16 shows $|B|$ along the longitudinal axis of the lens (zeroed on the centre on the lens) for $(x, y) = (7.75, 0)$ mm (the edge of the borehole). This is the maximal flux density inside the lens. The analytical approach assumes infinitely long segments when determining the analytic form of $|B|$. In the simulation of section 5.3.1, $|B| = 0$ outside of the lens borehole. The FEM uses segments of finite length and models the flux density outside the lens's end - seen as tails in figure 5.16.

Figure 5.17 shows two interpolated transverse cuts of the norm of the magnetic field, $|B|$, in the lens. Cut a), taken in the longitudinal centre of the lens, shows the quadratic flux density that we expected from the analytical model. Cut b), taken at the end of the lens, show quadratic flux density but at a much lower magnitude. Decreased $|B|$ will result in decreased ω .

Quadratic fits were applied to $|B|$ in the transverse y -axis. The coefficient, c , of the second-degree term was then compared with the prediction from the analytical model. From equation 5.8, we expect the following relationship,

$$c = \frac{2B_3}{\rho_1^2}. \quad (5.65)$$

For our specific lens design, this results in $c = 29.5 \text{ kT/m}^2$. Figure 5.18 illustrates the variation of c as a function of longitudinal position, with the centre of the lens as the reference point (zero position). The analytical prediction is represented by the dashed blue line, which is expected to remain constant inside the lens and zero outside. The solid black line corresponds to the prediction from the finite element model (FEM).

To determine the FEM value of c inside the lens, the central 50% of the data within the lens were averaged, excluding the outermost 25% from each end where the value began to decrease. The resulting mean value was $c_{\text{mean}} = 27.7 \text{ kT/m}^2$, which is approximately 6% lower than the analytical prediction. Towards the edge of the lens, c decreases to 50% of c_{mean} , and further decreases to 10% at a distance of 0.5 cm outside the lens.

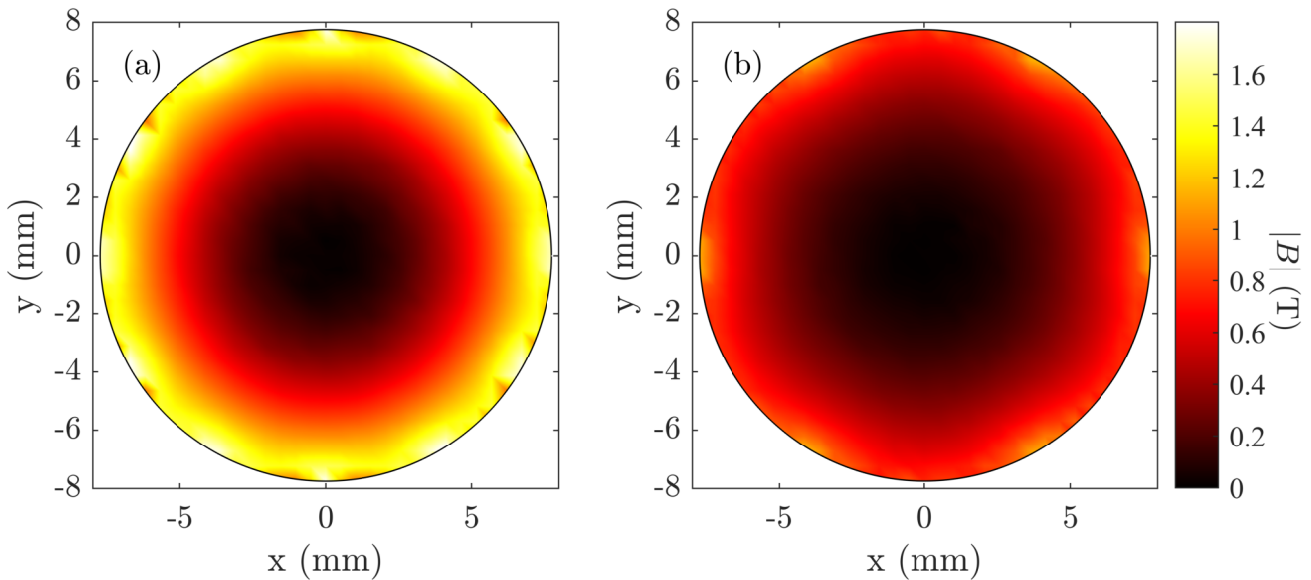


Figure 5.17: Simulated transverse cuts of the norm of the magnetic field, $|B|$, in the bore of the lens. a) is in the centre of the lens. b) is at the end of the lens. Note that the simulated data were interpolated.

The mean value c_{mean} was incorporated into the analytical model. Although it resulted in a slight reduction in the lens frequency, the overall effect was negligible. The main impact was observed in the range of molecular longitudinal velocities that the lens effectively guides. However, this shift can be easily compensated by making a small adjustment to the longitudinal position of the lens.

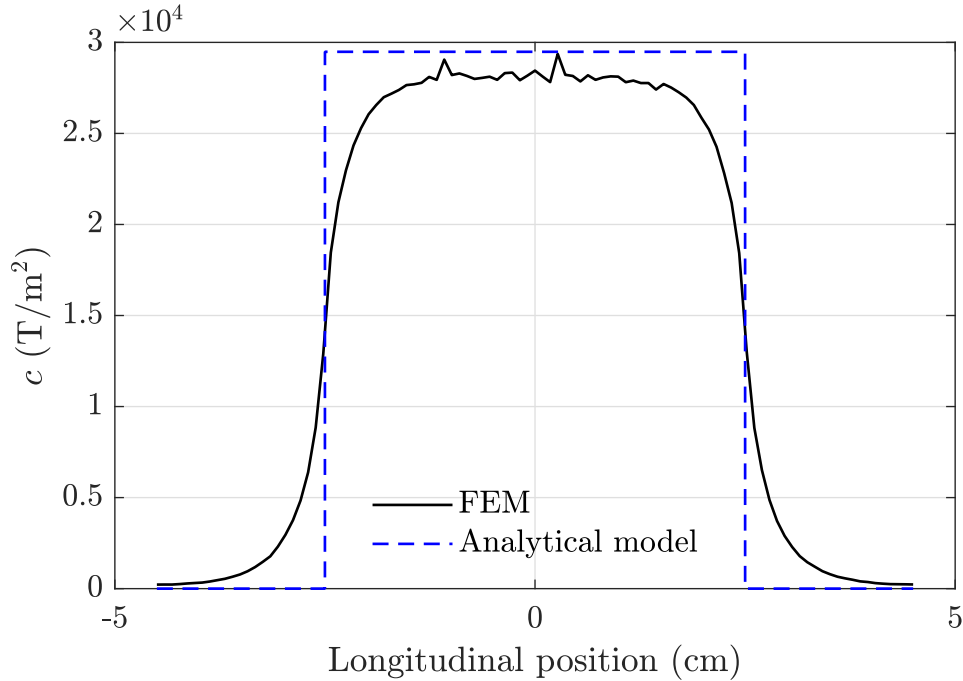


Figure 5.18: Simulation of the second-degree term coefficient, c , as a function of longitudinal position (zeroed on the centre of the lens). The black line shows the FEM linear model best-fit results. The dashed blue line shows the analytical model prediction.

Compared to the full length 15 cm lens, the tails of c span a short longitudinal range. It is assumed that when the three 5 cm long sections are assembled, the flux density at the joining points will be equivalent to the centre of the 5 cm model discussed here. A FEM of the full lens was not performed because tripling the number of components would have forced a reduction in model resolution unless a more powerful computer was used. A poorer resolution than the one used here would fail to capture the effects of the filleted edges.

5.3.3 Improvement on current scheme

Consider the case in which there is no magnetic lens present. A molecular source produces YbF molecules with initial ellipse parameters $(\gamma_0, \alpha_0, \beta_0)$. They freely evolve across the 57 cm distance

between the source and cooling lasers, taking time t_1 to traverse the gap. From section 5.2.4 we find that the molecules evolve as,

$$\begin{pmatrix} \gamma \\ \alpha \\ \beta \end{pmatrix} = M_{\text{Free}}(t_1) \cdot \begin{pmatrix} \gamma_0 \\ \alpha_0 \\ \beta_0 \end{pmatrix} \quad (5.66)$$

In this case, only 29% of the laser cooling acceptance (LCA) overlaps with the molecular phase space distribution, but this is true for only one of the transverse axes. When squared, this corresponds to only 8.4% of the four-dimensional LCA being overlapped. However, depending on the longitudinal velocity of the molecular beam and the positioning of the lens, the lens design presented in this work achieves O_{LCA} ranging from 95% to 100%. When squared to account for all four dimensions, this translates to an overlap of 90% to 100% in 4D phase space. Consequently, the lens design provides a substantial increase of 10.7 to 11.9-fold in the overlap with the 4D LCA.

It is important to note that the molecules will be evenly distributed between the two valence electron spin states, resulting in half of them being guided and the other half being anti-guided by the lens. Therefore, we can expect 5.3 to 6.0 times more molecules to be laser cooled compared to the initial distribution. It is worth considering that molecules could be optically pumped into the weak-field-seeking states before entering the lens, allowing all of them to be guided. This approach would double the number of ultracold molecules at the expense of introducing further experimental complexity.

The brightness of our ultracold YbF beam, running at a repetition rate of 10 Hz, was measured to be greater than 1×10^{17} molecules/m²/ster/s (see section 4.4.5, [1]). If the number of laser-cooled molecules increases by a factor of 5.3, then so will the brightness of the ultracold molecular beam - resulting in a beam of brightness greater than 5.3×10^{17} molecules/m²/ster/s.

Finally, let's consider the improvement this could make to the precision of an eEDM measurement. Extrapolating from the estimate for statistical uncertainty in section 4.4.6, where we found that

after 100 days (a typical length of time for an eEDM measurement), we could expect a statistical uncertainty of as little as $\sigma_{d_e} = 5 \times 10^{-31}$ e cm [1]. Taking a maximally pessimistic approach, we could expect a 5.3-fold increase in the number of detected molecules with the addition of the magnetic lens. This would reduce the statistical uncertainty by a factor of 2.3. Therefore, after a 100-day measurement, we would expect $\sigma_{d_e} = 2.2 \times 10^{-31}$ e cm. Such a measurement would be a factor of 18 more precise than the JILA result, which sets the current best limit [16].

5.4 Magnetic lens design

5.4.1 Design

Magnet segments

Figure 5.19 shows a cross-section and an angled view of a magnet segment. The inner and outer radii are 7.75 mm and 37.25 mm respectively. With a segment angle of 30° , 12 of these complete the Halbach cylinder. The length of 50 mm is a manufacturing constraint. Similarly, the 0.5 mm filleted edges are to reduce the risk of accidental damage to the brittle material.

An important design feature is the azimuthal trimming of the segments. When mounting a magnet segment, the inner radius must be flush with the inner surface of the mount. If one does not account for the spoke widths, the segments will be radially offset from the mount centre - increasing the bore radius of the lens and decreasing the lens frequency. This is avoided by removing a half-spoke width plane from each side of the segment - creating a 0.25 mm gap between segments when assembled in the mount. This design feature was included in the FEM results presented in section 5.3.2.

Figure 5.20 shows the magnetisation directions of the magnets in this 12-segment design. It requires segments with only two distinct magnetisations, E and B, angled $\pm 45^\circ$ relative to the horizontal dashed line. Rotating these segments about the longitudinal axis results in styles E* and B*. The cylinder is assembled with the repeating pattern B*E*EB (or any circular shift permutation

of these elements). The magnetic remanence of the magnets is quoted from the manufacturer at 1.44 T - the strongest they can make.

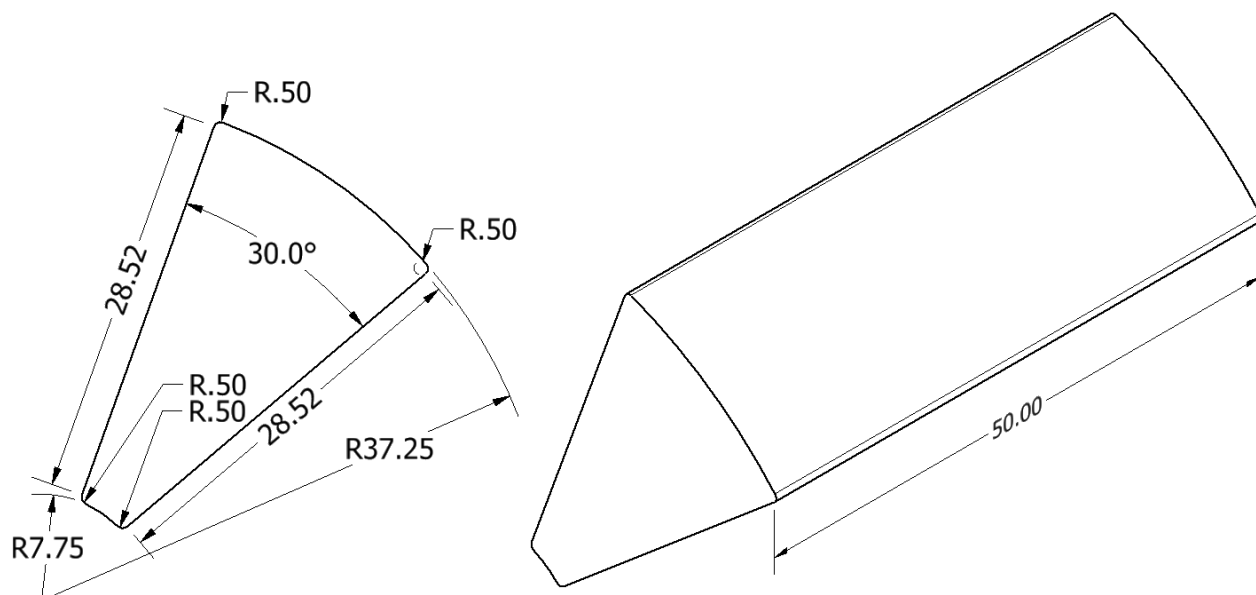


Figure 5.19: Cross section and angled view of a magnet segment. All annotated dimensions are written in millimetres.

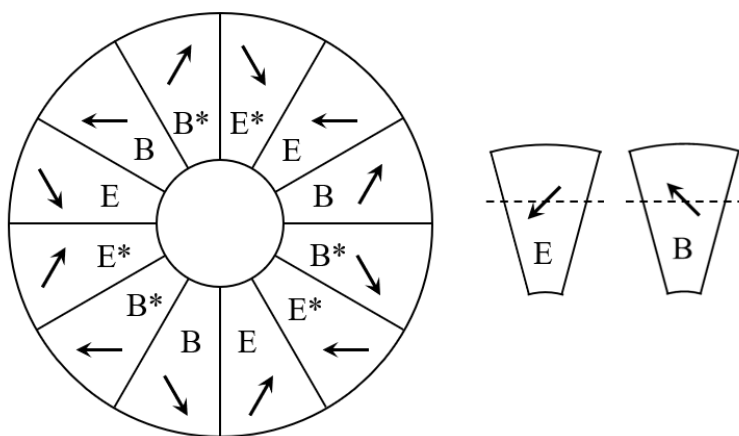


Figure 5.20: magnetisation directions of the magnet segments. This 12-segment design requires only two styles of magnetisation, E and B, with $\pm 45^\circ$ between them and the horizontal dashed lines. Rotation of E and B about the longitudinal axis results in E* and B*.

Magnet mounts

Figure 5.21 shows a schematic of a magnet mount. A 10 mm thick, 46.25 mm radius, stainless steel disk has a 7.25 mm radius disk cut out of the centre. 30° segments are cut out, leaving 0.25 mm

spokes and an inner ring of radial thickness 0.5 mm. The remaining outer ring has inner radius 38.25 mm and radial thickness 8 mm. Six uniformly spaced 2 mm radius holes are drilled through the outer ring. These will house supporting stainless steel rods.

The angled view in figure 5.21 shows radially tapped holes. For each magnet segment, there is a radially tapped M4 through-hole. M4 grub screws will be used to mechanically hold the magnets in the mount. For each of the 2 mm radius holes in the cross-section, there is an M3 radially tapped hole. M3 grub screws will mechanically hold the stainless steel rods in place.

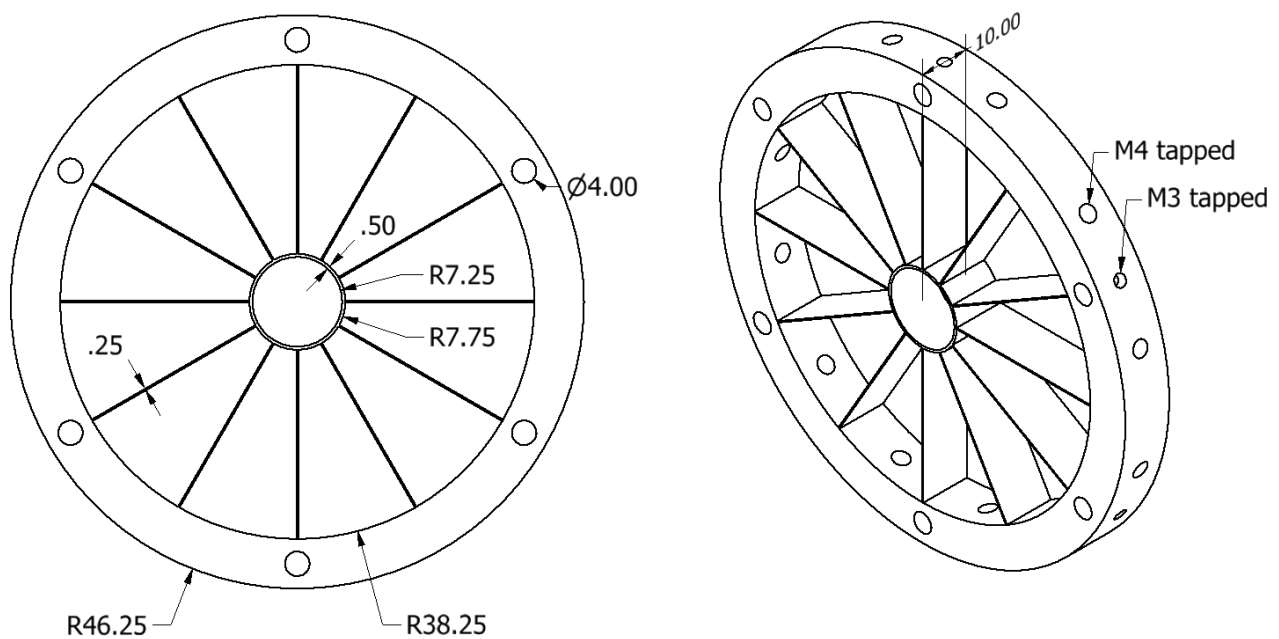


Figure 5.21: Cross section and angled view of a magnet mount. All annotated dimensions are written in millimetres.

Full lens assembly

Figure 5.22 shows a model of the 5 cm magnet segments installed in three mounts - a stainless steel mount at each end of the segments and an aluminium mount in the centre to guide them during assembly. Not shown are the grub screws, which will mechanically hold the magnets in the mounts. Also not shown are the stainless steel rods which connect the mounts to one another.

The model of the full lens assembly is shown in figure 5.23. Stainless steel rods of diameter 4 mm connect the three 5 cm long sections to complete the 15 cm magnetic lens. Two aluminium fix-

tures connect to the rods at opposite ends and secure the lens to the inner surface of the vacuum chamber via grub screws.

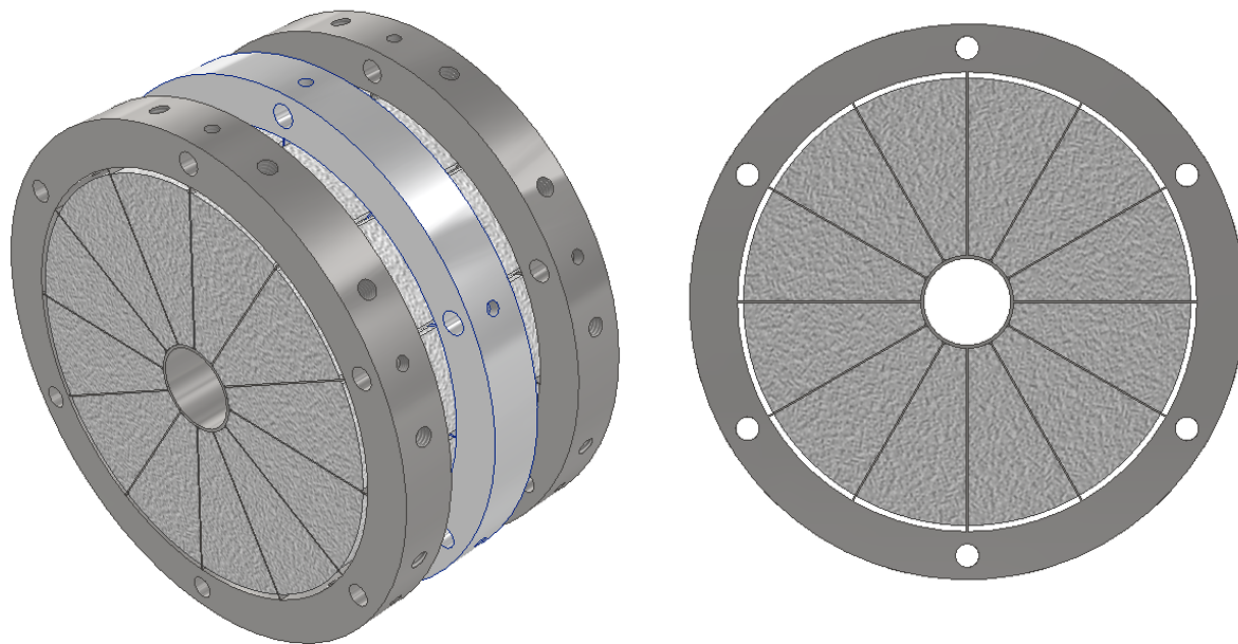


Figure 5.22: 5 cm long magnet segments installed in three mounts: two stainless steel and an aluminium mount for guiding the segments during assembly. Stainless steel rods and grub screws are not shown.

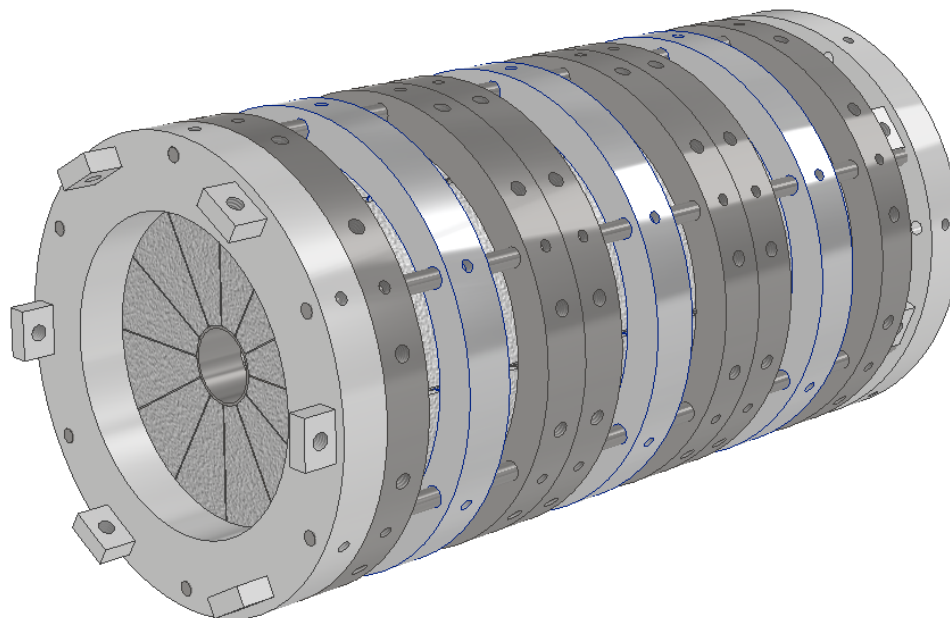


Figure 5.23: Full lens assembly. Three 5 cm sections (see figure 5.22) are connected by six stainless steel rods. At each end there is an aluminium part which mounts the lens inside the vacuum chamber. No grub screws are included in this schematic.

5.4.2 Building the lens

Assembling the magnetic lens was difficult due to the strong forces between magnet segments. Once all segments are mounted, the internal forces oppose one another, and the mount spokes are not under much stress. However, whilst assembling the lens, the spokes must endure very strong forces. Initial attempts to use aluminium magnet mounts failed as the spokes separating the segments broke before all segments could be mounted. This was mitigated using three measures: the mounts at the ends of each 5 cm section were manufactured from stainless steel (much stronger than aluminium); dummy magnet segments made of aluminium were used in the empty sections during assembly to distribute the forces between more spokes and finally, additional magnet mounts, made of aluminium, were added in between the stainless steel mounts to better distribute the forces and to help guide the magnet segments into their final positions.

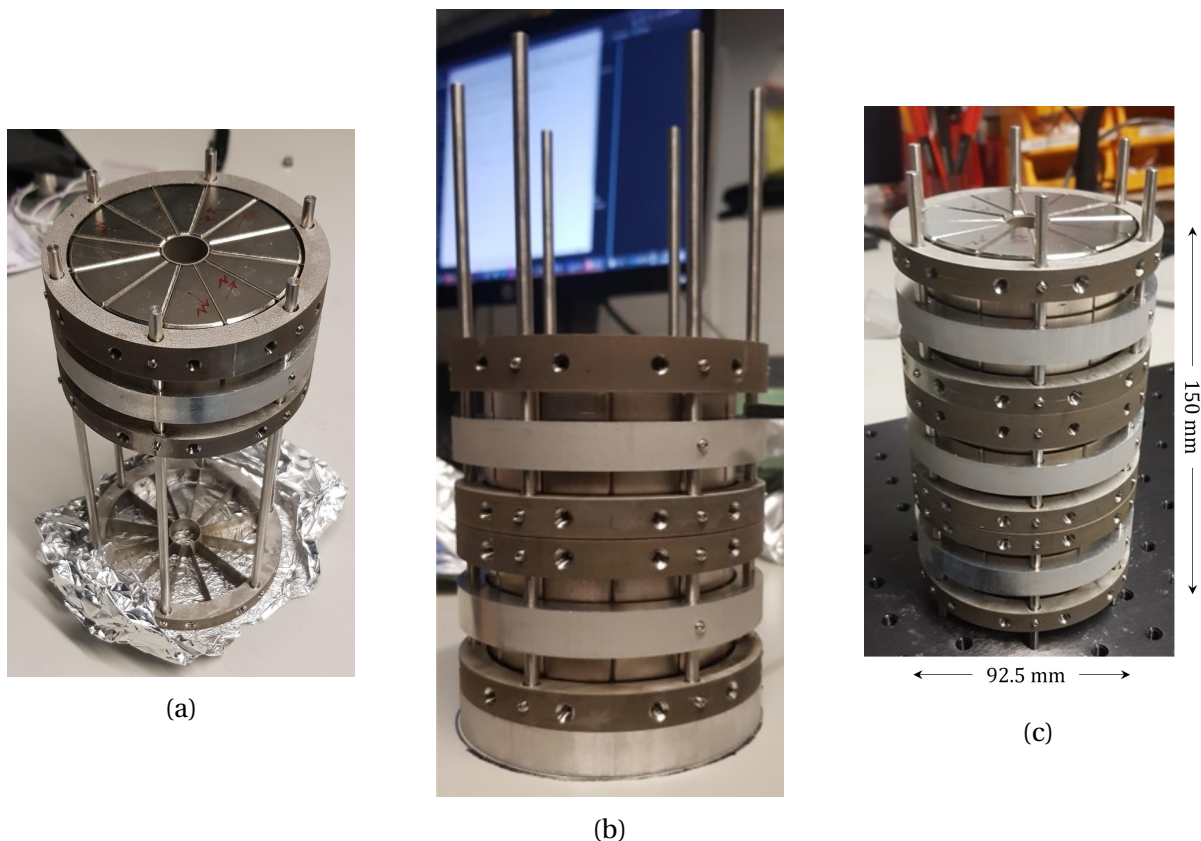


Figure 5.24: Lens during assembly. Photos (a)-(c) show the lens at varying stages of completion - having one, two and three of the 5 cm sections assembled.

Figure 5.24 shows the magnetic lens at varying stages of assembly. It was found that the longitudi-

nal forces between the 5 cm sections were quite large. As such, a clamp had to be used to compress it before tightening the grub screws to the connecting rods.

5.4.3 Characterization of the lens

Radial field strength outside the lens

A measurement of the radial field outside the lens was performed and compared with the finite element model discussed in section 5.3.2. The lens was stood on its end and rotated in increments of 7.5° whilst the radial magnetic field was measured using a gaussmeter. Figure 5.25a depicts the coordinate frame used for this measurement whilst figure 5.25b shows the transverse magnetic field calculated using a finite element model in COMSOL.

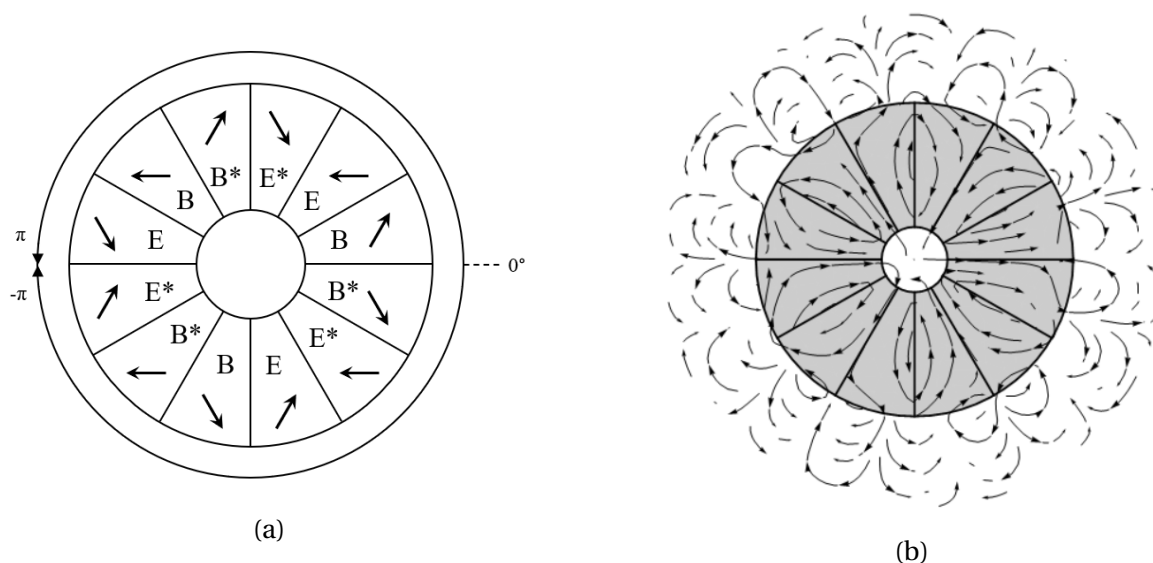


Figure 5.25: (a) Coordinate frame used to measure radial field as a function of azimuthal angle. (b) Simulated stream plot of the transverse magnetic field, calculated using a finite element model in COMSOL. Overlaid in grey are the magnet segments.

Precise positioning the gaussmeter probe was challenging. As such, I assumed a large uncertainty in the angle of the probe. Figure 5.26 shows the FEM prediction alongside the measured values. The two data sets agree well, considering the difficulties faced when accurately positioning the probe in a field with such steep gradients.

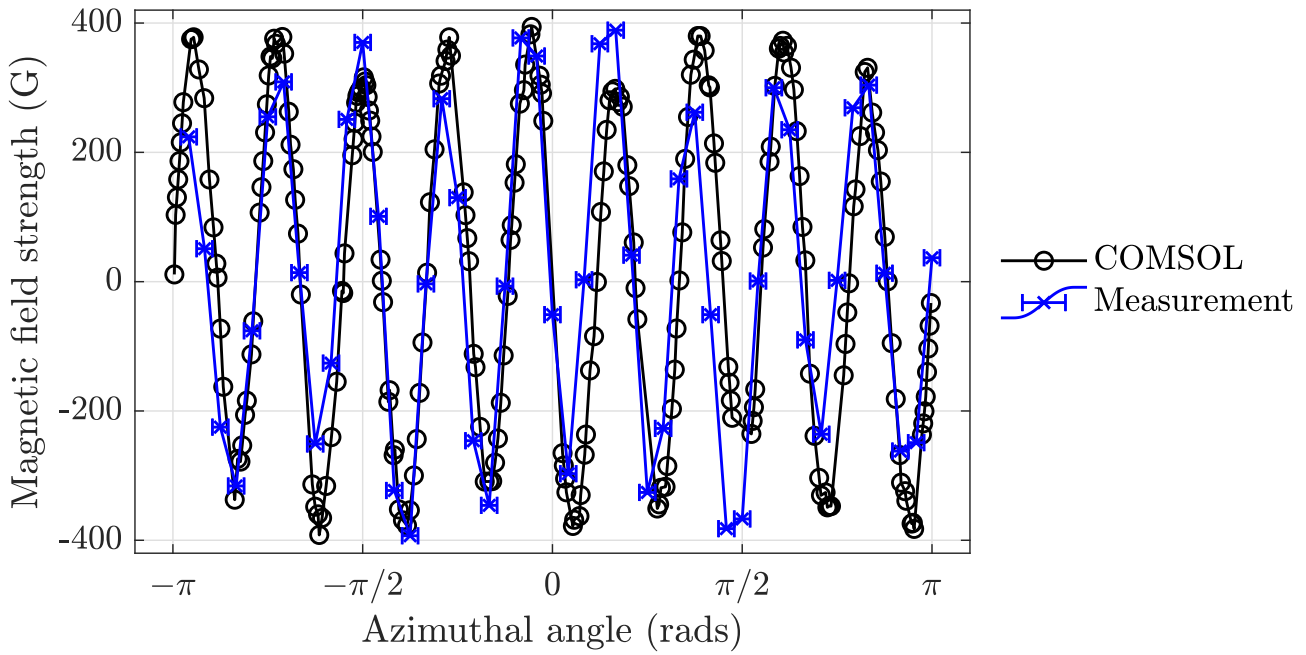


Figure 5.26: Radial magnetic field strength 13 mm from the outside edge of the cylinder. Black circles show the simulated finite element model prediction made using COMSOL, whilst the blue crosses show measurements made using a Gaussmeter.

Radial field strength inside the lens

The radial field strength inside the bore of the lens was measured as a function of transverse position. A gaussmeter probe was mounted on a translation stage with the tip of the probe placed approximately 2 cm inside the cylinder and facing in the radial direction. The translation stage was then used to move the probe in increments of 0.64 mm from the centre of the bore to as close as possible to the inner surface of the magnets - limited, of course, by the inner rings of the magnet mounts.

Figure 5.27 presents both the measurement and the prediction obtained from COMSOL. A noticeable difference between the two is observed, which was not unexpected given the difficulty in accurately positioning the probe. However, a more promising insight is obtained by applying the quadratic fit $a + c(\rho - \rho_0)^2$, where $\rho_0 = 0.312(35)$ mm and $a = -256(18)$ G indicate a systematic offset in the measurement from the centre. In fact, translating the data leads to a visibly improved agreement with the simulation. Nonetheless, the obtained value of $c = 24.25(31)$ kT/m² is approximately 12% lower than the FEM prediction. It is hoped that adjusting the longitudinal position of

the lens will compensate for this reduced lens frequency.

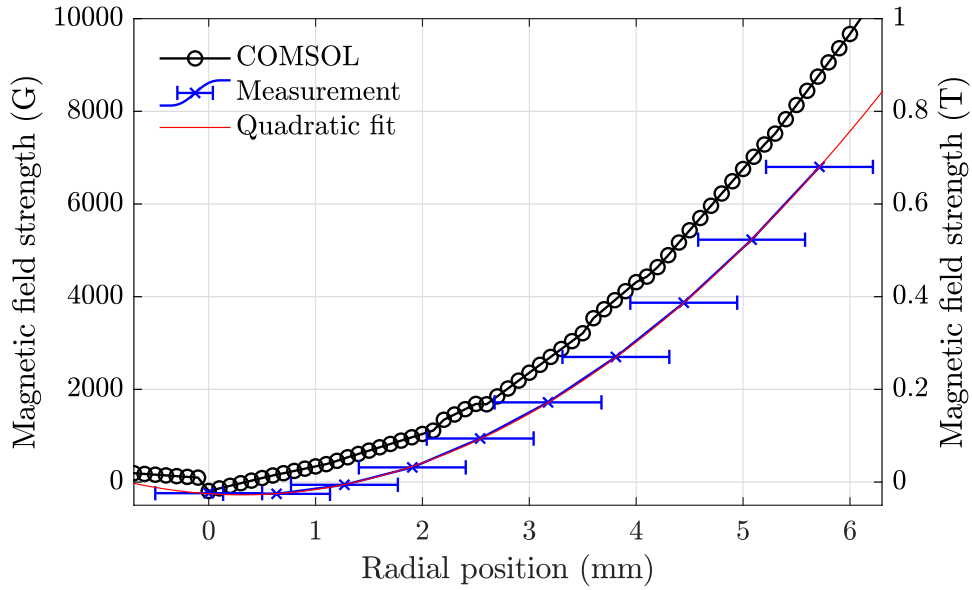


Figure 5.27: Radial magnetic field strength inside the cylinder. Field strength is provided in Gauss and Tesla for the reader's convenience. Black circles show the simulated finite element model prediction made using COMSOL, whilst the blue crosses show measurements made using a Gauss-meter.

5.5 Summary

In this chapter, I have presented the design, construction, and characterisation of a magnetic lens specifically developed for the YbF molecular funnel. The foundation of the physical model relies on both an analytical framework proposed by Halbach [108] and a detailed finite element model. The simulation techniques employed draw inspiration from those commonly used in high-energy particle physics. The lens has been successfully constructed and is currently awaiting installation in the YbF beamline.

Measurements of the magnetic fields inside and outside the lens have been carried out to evaluate its performance. The results obtained outside the lens are in excellent agreement with the predictions from my simulations, confirming the accuracy of the model. However, the measurements conducted inside the lens reveal a discrepancy, with gradients found to be 12% smaller than initially expected. This discrepancy may be attributed to the inherent challenges of accurately mea-

suring large field gradients within a confined space.

Nonetheless, the simulations of the lens demonstrate its capability to effectively guide molecules with a wide range of longitudinal velocities towards the laser cooling region. By adjusting the longitudinal position of the lens, it becomes feasible to finely tune the range of longitudinal velocities that are efficiently guided. This flexibility allows for the optimisation of the lens performance to match the specific requirements of the experiment, accommodating both higher and lower longitudinal velocity ranges as desired.

Simulations have demonstrated that the combination of a magnetic lens with transverse laser cooling can enhance the brightness of our molecular beam by a factor of 5.3. This enhancement has significant implications for precision measurements of the electric dipole moment in YbF. By reducing the statistical uncertainty by a factor of 2.3, using the magnetic lens can substantially improve the precision of an eEDM measurement. Such a measurement would be a factor of 18 more precise than the JILA result, which sets the current best limit [16].

Chapter 6

Transferring YbF molecules to the eEDM measurement state

Laser cooling YbF molecules requires the molecules to be in the first excited rotational state, $N = 1$, as discussed in chapters 2 and 4. However, the eEDM measurement is performed on molecules in the superposition state $|N, F, m_F\rangle = |0, 1, -1\rangle + |0, 1, +1\rangle$. Therefore, after laser cooling, the molecules must be transferred from $N = 1$ to $N = 0$. This chapter discusses the development of a new scheme for transferring molecules efficiently between these rotational states.

6.1 Optical pumping and rotational state microwave mixing

The pumping scheme for transferring molecules from the laser cooling state to the ground rotational state uses a mixture of microwave and optical transitions. Figure 6.1 shows the transitions involved. Blue lines depict the microwave coupling of the first and second excited rotational states, and green lines show the optical transitions between the ground and excited electronic states. The laser-cooled molecules start in the $N = 1$ rotational state. Continuous microwave radiation applied to the molecules induces Rabi oscillations between $N = 1$ and $N = 2$. Laser light, used concurrently with the microwaves, pumps the molecules from $N = 2$ to the excited electronic state $A^2\Pi_{1/2}(\nu' = 0, J' = 1/2)$, where they then decay predominantly to $N = 0$. Additional laser

light addresses the $X^2\Sigma^+$ ($N = 0$, $F = 1$) state, such that other than the higher vibrational states, the molecules are all pumped into $X^2\Sigma^+$ ($N = 0$, $F = 0$). In the future, the molecules will then be transferred from $X^2\Sigma^+$ ($N = 0$, $F = 0$) into the superposition state used for probing the eEDM.

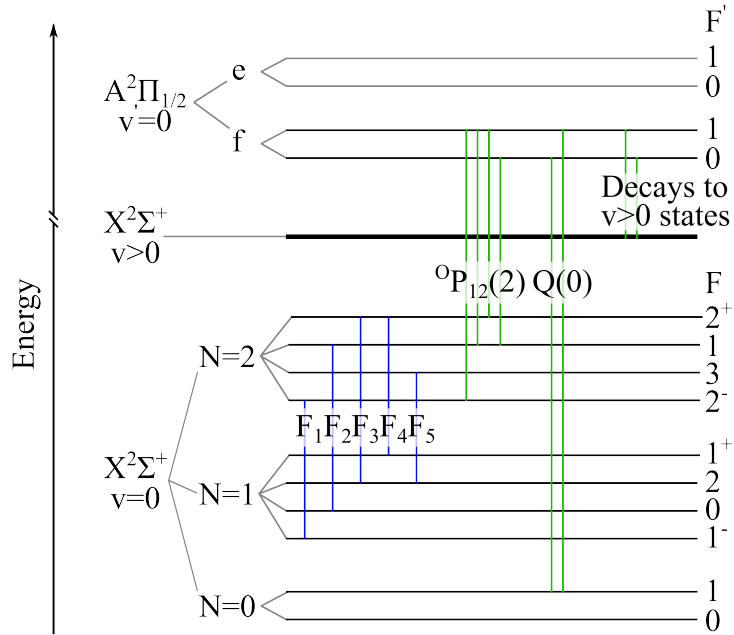


Figure 6.1: Transitions involved in transferring molecules from the laser cooling state to the eEDM measurement state. Microwave transitions are shown by blue lines, connecting the first and second excited rotational states of $X^2\Sigma^+$ ($v = 0$). Green lines show optical transitions between $X^2\Sigma^+$ and $A^2\Pi_{1/2}$ ($v' = 0$) states.

All optical transitions used for this pumping scheme are the same as those used in a previous YbF eEDM experiment [3, 41]. However, I present a new approach for microwaves, coupling $N = 1$ and $N = 2$ instead of $N = 1$ and $N = 0$. This removes the need for RF fields and eliminates the dark states that would otherwise exist. Another advantage is that the microwaves used for this pumping scheme will not affect molecules in the $N = 0$ eEDM measurement state.

In figure 6.1, the first four microwave couplings, $F_1 - F_4$, connect all of the $N = 1$ hyperfine states to $N = 2$. The frequencies required to address F_1 and F_4 only differ by 1.8 MHz and can be addressed with a single frequency, provided the Rabi frequency is comparable to, or larger than, this splitting. Similarly, F_2 and F_3 differ by 11 MHz, and since the weak F_3 transition requires a lot of power to be addressed, the F_2 transition is off-resonantly driven. These two frequencies address the coupling of all four $N = 1$ hyperfine states to $N = 2$. However, the frequency used to address F_1 and F_4 differs by only 8 MHz from transition F_5 . As a result, a small part of the population is driven to ($N = 2$, $F =$

3), which cannot be coupled to the electronically excited state by the laser light. To solve this, a third microwave frequency is required to address the otherwise dark state $X^2\Sigma^+(N=0, F=3)$. Due to equipment failure, this third frequency, F_5 , was not tested experimentally, but simulations by Joanna Zielinska suggest that it is a necessity for efficient pumping. Nevertheless, the results I present are very promising.

6.2 Experimental setup

Chapter 3 provides an overview of the experimental setup for the YbF experiments. The purpose of this section is to describe the infrastructure and techniques that are specific to this chapter, including the laser and microwave systems. First, section 6.2.1 provides an overview of the apparatus, followed by detailed discussions of the pumping region in section 6.2.2 and the detection region in section 6.2.3. Finally, section 6.2.4 outlines the measurement sequence and analysis methods, which differ from those used in the laser cooling experiments previously discussed.

6.2.1 Overview of the YbF pumping apparatus

The new YbF pumping scheme was developed on a shorter beamline than the one depicted in section 3.1, figure 3.1, as the interaction region, (6), and eEDM measurement state preparation regions (5) and (7), were not yet built. Figure 6.2 shows the beamline used; without the laser cooling region and (relocated) cleanup region depicted. Whilst laser cooling and the pumping scheme will eventually be used together, this work focussed only on developing the pumping scheme without the application of laser cooling. The molecules freely propagated between the source and LIF detector, with pumping taking place 88 cm from the source. Pumping light propagates into and out of the page in the $\pm\hat{x}$ directions, whilst microwaves are emitted by the horns mounted above the molecular beam.

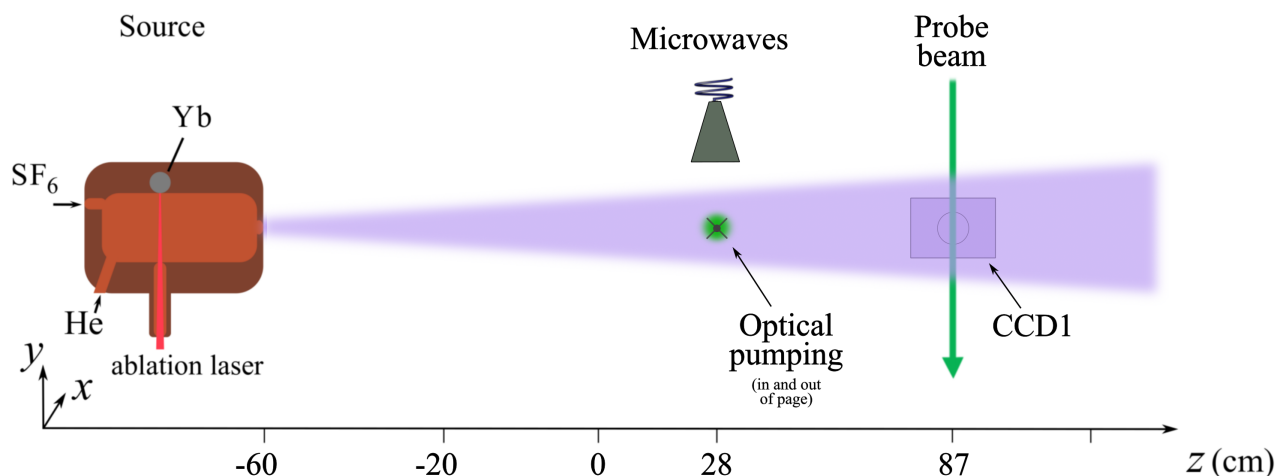


Figure 6.2: Diagram of the YbF pumping experiment. The laser cooling region, whilst unused, was still situated between $z = -20$ cm and $z = 0$ cm, as in figure 4.2. The molecules travel through optical pumping light in the presence of microwaves emitted by microwave horns. After propagating freely for 58 cm, the molecules are imaged using an EMCCD. Figure adapted from Alauze et al. (2021) [1].

6.2.2 Pumping region

The only significant change to the beamline used in the laser cooling experiment (section 4.2.1) was the chamber used for pumping. Figure 6.3 shows a CAD drawing of the beamline chamber where optical pumping occurred. The molecular beam travelled from right to left through the chamber. At the top of the figure, two Pasternack PE9850/SF-20 microwave horns were mounted inside a vacuum flange. Each was fixed to a rotatable mount pointed towards $(x, y) = (0, 0)$, the centre of the molecular beam. Two smaller flanges, affixed to the same flange as the horns, contained two hermetically sealed microwave feedthroughs (Fairview Microwave FMAD1201) to transmit microwaves into the vacuum chamber to the horns. On each side of the chamber, rows of windows provided optical access for laser cooling clean-up light and optical pumping light.

The laser light for optical pumping was delivered to the experiment by optical fibres. Fast shutters, situated before the optical fibres, blocked the pump light when needed. Figure 6.3 shows the optical pumping light reaching the experiment and addressing the molecular beam from the $\pm \hat{x}$ directions. The pump beam was collimated with a waist of $w = 9.8$ mm using a fibre collimation pack-

age¹, where the waist was defined by the Gaussian intensity profile of the beam $I(r) = I(0)e^{-2r^2/w^2}$.

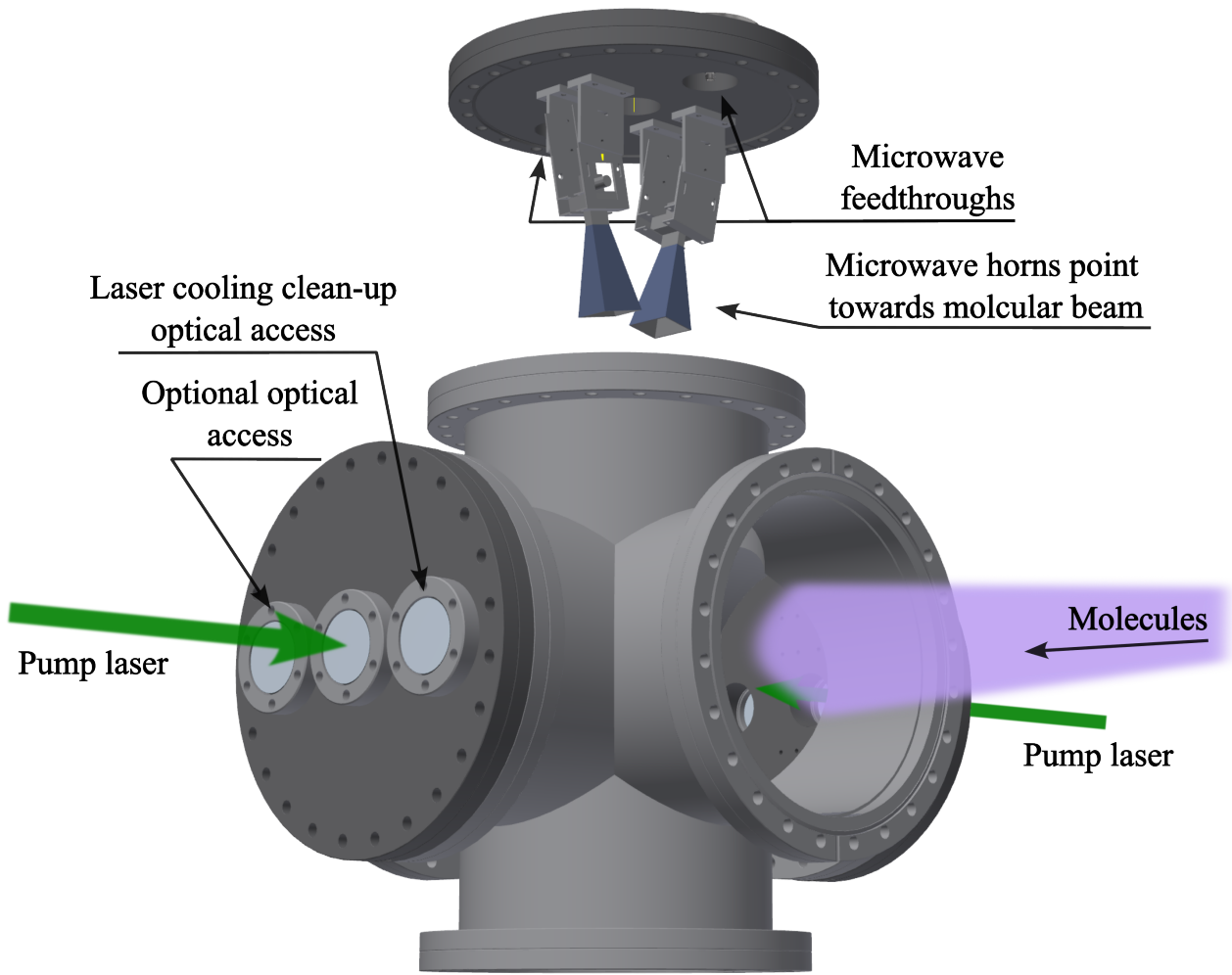


Figure 6.3: CAD drawing illustrating the molecule pumping region, showing the path of molecules as they travel down the beamline and interact with pumping lasers in the presence of microwave fields. Microwaves were emitted by microwave horns mounted inside a vacuum flange.

6.2.3 Detection

Light-induced fluorescence, captured by an EMCCD camera, facilitated the detection of molecules for all measurements in this chapter. The same technique was used for the laser cooling experiments presented in chapter 4. However, different probe light frequencies were used for the experiments discussed in this chapter, depending on the independent variable being studied. For exam-

¹The Thorlabs fibre collimation package (F240APC-A) was specified to output a beam of $1/e^2$ waist 1.5 mm. However, combined with our choice of fibre, the resulting beam waist was much larger and comparable to the transverse width of the laser-cooled molecular beam.

ple, when studying how efficiently the molecules could be optically pumped out of $X^2\Sigma^+(N=2)$, single-frequency probe light was applied to induce fluorescence in each of the $N=2$ hyperfine states.

Figure 6.4 shows two example images from the molecular pumping experiments. On the right, unpumped molecules in $X^2\Sigma^+(N=1, F=1^+)$, travelling in the $+\hat{z}$ direction, fluoresced in the probe laser light. In the left-hand image, pumping was applied to the molecular beam. Many molecules were pumped out of the target state, but some unpumped molecules are seen at the top and bottom; these molecules passed through the wings of the Gaussian pump beam and were not efficiently pumped. The red rectangles show the region of interest for analyses presented in this chapter, with the wings of the probe beam ($\pm\hat{z}$ directions) and the inefficiently pumped molecules ($\pm\hat{y}$ directions) cropped for accurate assessment of pumping efficiency.

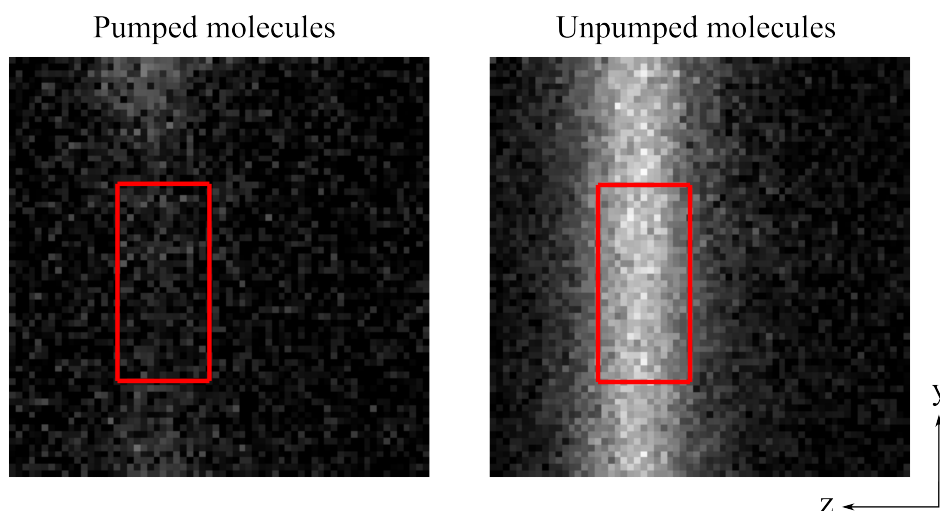


Figure 6.4: Example images from the EMCCD camera, with the same greyscale contrast settings. Both are averages of 99 photos with the averaged background subtracted. On the left, molecules have been pumped out of the laser cooling state. The right-hand image is of the unpumped molecular beam. Red rectangles denote the region of interest for our analysis. The pixel scale was not calibrated in this camera position, as it was unnecessary for optimising pumping.

6.2.4 Measurement sequence and analysis

The measurement sequence used for the pumping experiments bore a lot of similarity to those used in the laser cooling experiments (see section 4.2.4). However, instead of turning laser cooling

on and off between measurements, molecular pumping was turned on and off. For each measurement, a set of four photos were taken with different experimental conditions:

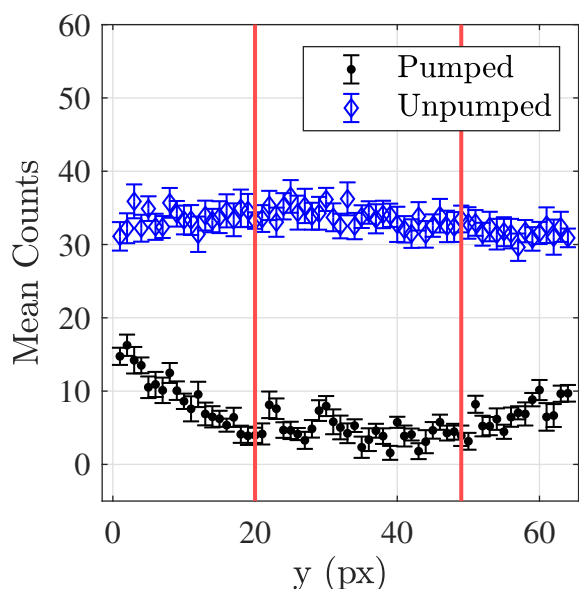
1. Molecules were created and pumped
2. No molecules were created (background photo)
3. Molecules were created, but neither pumping light nor microwaves were applied
4. No molecules were created (background photo)

Photos of pumped and unpumped molecules were taken with background photos for each. The pumping light was shuttered before photos were taken to reduce background light. Photo 2 was a background image taken immediately after photo 1. Similarly, photo 4 was a background image for photo 3. This set of four photos was the basis for most measurements in this chapter. Photos were taken in this order repeatedly until sufficient data were available for averaging to achieve the desired image quality. Depending on the number of molecules produced by the source, which could vary considerably daily, we averaged 150–400 photo sets².

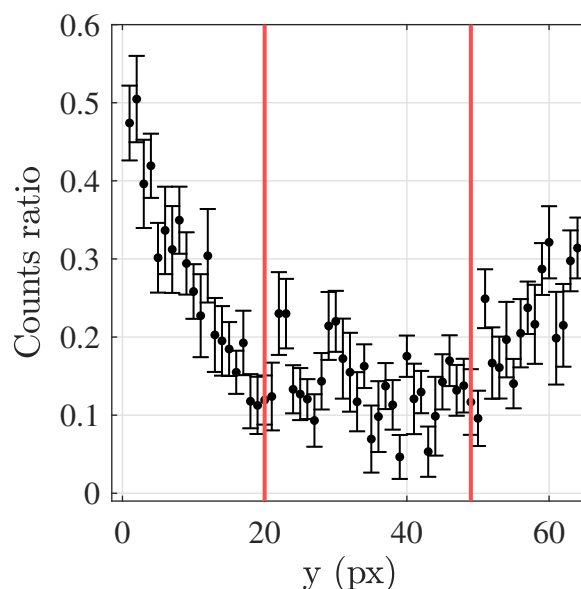
To analyse the data, photo sets were first averaged, producing the four images above. After subtracting the background images, they were cropped to the region of interest. The ratio, R_i , of pumped to unpumped mean counts was then calculated for each pixel i . The mean of these ratios, $\bar{R}$, was computed, and the uncertainty in $\bar{R}$ was determined by propagating the standard error from the pixel count means.

Figures 6.5a and 6.5c show averaged cross-sections of the images in figure 6.4. The vertical red lines correspond to the red rectangles overlaid on the images. In figure 6.5a, partially pumped molecules are seen in the regions $y \lesssim 20$ px and $y \gtrsim 49$ px. This is clearer in figure 6.5b, where the ratio of the two curves is shown. Observation of partially pumped molecules was expected because the pump beam had a finite waist of 9.8 mm in the y -axis. Thus, the analysis only considered molecules in the region $20 \text{ px} < y < 49 \text{ px}$.

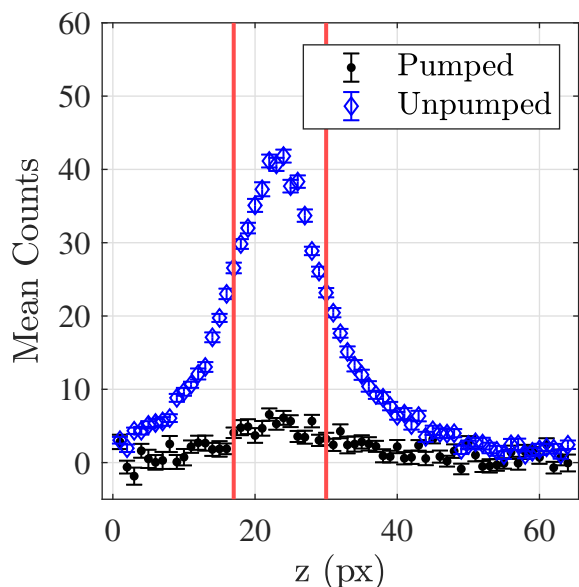
²The number of photos taken for the pumping experiments was 1.5–2.5 times less than the laser cooling experiments because the camera was closer to the source, meaning that the molecular signal was stronger.



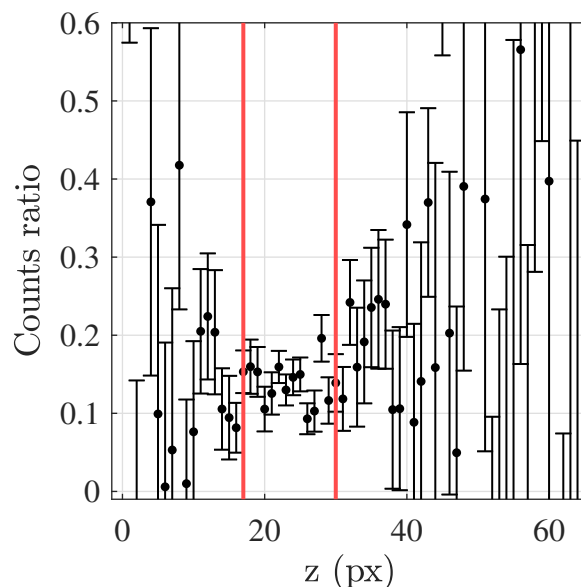
(a) Mean pixel counts as a function of y position. The mean was taken for points between $z = 17$ px and $z = 30$ px - the red lines in plot (c).



(b) Ratio of the curves in plot (a). Data in the regions $y < 20$ px and $y > 49$ px are increasing in magnitude because of the finite width of the pump beam.



(c) Mean pixel counts as a function of z position. The mean was taken for points between $y = 20$ px and $y = 49$ px - the red lines in plot (a).



(d) Ratio of the curves in plot (c). Data in the regions $z < 17$ px and $z > 30$ px are discarded because they are outside of the probe beam.

Figure 6.5: Examples of mean LIF counts on the EMCCD camera, using the same data as in figure 6.4. Plots (a) and (c) show the mean pixel counts; blue diamonds show the unpumped molecules, while black points show the pumped molecules. The vertical red lines show the region of interest for analyses in the chapter. Plots (b) and (d) show the ratios of the curves in plots (a) and (c).

Figure 6.5c shows mean camera counts against the z -axis. Molecules were only visible to the camera when illuminated by the probe light, so, in this axis, both curves traced the Gaussian profile

of the probe beam. Figure 6.5d shows the ratio of the curves in figure 6.5c. Outside of the probe beam, the pixel ratios had large variations and statistical noise. Therefore, this data was discarded from the analysis - with only those pixels in the region $17\text{px} < z < 30\text{px}$ being used.

6.3 Results

In this section, the results of the pumping experiments are presented and discussed in detail. The findings are categorised into three main sections. Sections 6.3.1 and 6.3.2 investigate the depletion of the $X^2\Sigma^+ N = 1$ and $N = 2$ states, respectively. The experiments were conducted under varying conditions of laser power, microwave frequencies, and microwave powers. The results show the effects of each parameter on the depletion process, and the implications of these findings are discussed. In section 6.3.3, the accumulation and depletion of the two hyperfine states in $X^2\Sigma^+ N = 0$ are presented.

6.3.1 Depletion of the $N=1$ hyperfine states

Here, the depletion of the $X^2\Sigma^+ (N = 1)$ hyperfine states and the factors that influence the processes are explored. Firstly, I discuss the probe light used to observe the states, providing insight into the experimental setup. Then, I present the results of investigations and analyses of the dependency of the pumping process on several key parameters, including microwave powers, microwave frequencies, and total $^{\text{O}}\text{P}_{12}(2)$ pump power. The findings are discussed in detail, shedding light on the underlying mechanisms that govern the depletion of the $N = 1$ hyperfine states.

To observe the depletion of the $X^2\Sigma^+ (N = 1)$ hyperfine states, we initially used the Q(1) branch to measure the population. However, the depletion ratios obtained never decreased much below 0.3, leading us to suspect that nearby transitions could interfere with the depletion measurements. Further investigations revealed that neither the pump light/microwave powers nor the interaction time were limiting factors. Subsequently, we switched to using the R(1) branch for probing, significantly improving the depletion ratio. It is worth noting that, from spectrum measurements

performed by J. Almond (2017) [94], we know the Q(1) branch is congested, supporting the conclusion that it is unsuitable for accurately measuring the depletion ratio of the $X^2\Sigma^+$ ($N = 1$) hyperfine states.

Figure 6.6 presents an R(1) branch spectrum measurement. A function was fitted to the data: the sum of five Lorentzians $g(f) = b \frac{\gamma}{\pi(\gamma^2 + (f - f_0)^2)}$, where b was a multiplication factor, γ the full width at half maximum and f_0 the centre frequency. The red dashed lines indicate the centre frequencies of the transitions associated with the R(1) branch, with their corresponding ground states being $F = 1^+$, $2 + 0$ and 1^- , respectively, from left to right, with the $F = 2$ and $F = 0$ peaks overlapping one another. Additionally, two transitions of unknown origin are located between the $F = 1^-$ and $F = 2$ transitions. However, since these transitions are of small amplitude and width, their effect on the measurements presented in this chapter was minimal.

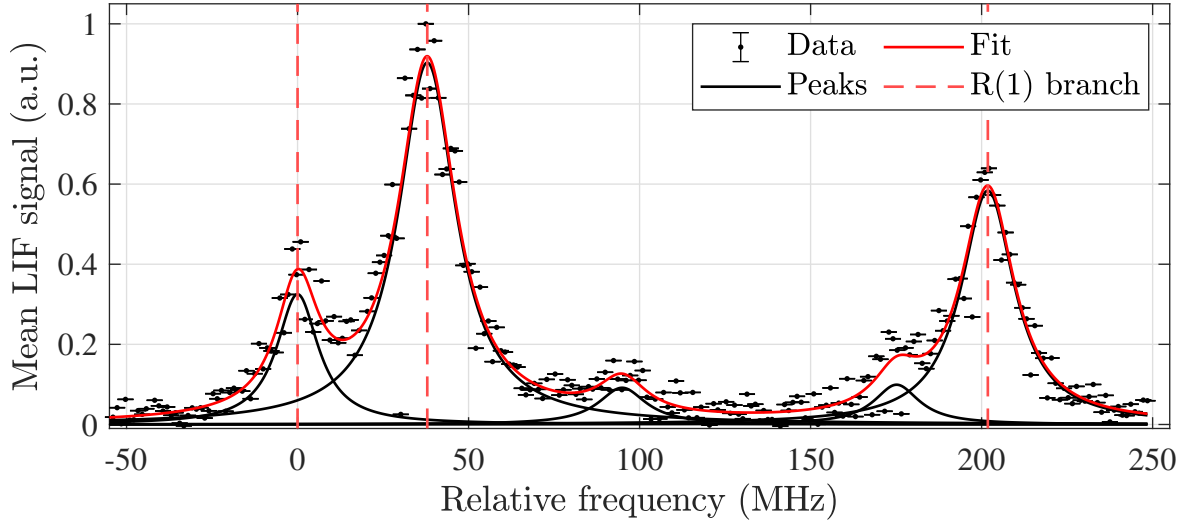


Figure 6.6: Spectrum for the $X^2\Sigma^+$ ($N = 1$) R(1) branch transitions, with the sum of five Lorentzian curves fitted. Data are shown as black points, with the fitted model overlaid as a red curve. The individual Lorentzian functions, $g(f)$, are plotted as black curves. Red dashed lines show the centre frequencies of the fits corresponding to R(1) transitions.

Dependency on $^0P_{12}(2)$ power

We needed to assess whether the laser power was adequate for our purposes. To do this, the depletion ratio of each $N = 1$ hyperfine state was measured using the R(1) transitions. The power of the $^0P_{12}(2)$ pump laser was varied between 6.6 mW and 45 mW, with $\sim 50\%$ of the power in the 185 MHz sideband. To ensure that the analysis focused solely on the $^0P_{12}(2)$ laser light, the Q(0)

pump light was kept off, thus avoiding any reduction in scattering rate that could occur with the inclusion of the Q(0) laser. Microwaves were applied at the maximum power output of 24.2 dBm at the input of the microwave horn.

Figure 6.7 displays the depletion ratio of the $X^2\Sigma^+$ ($N = 1$) hyperfine states $F = 1^-$, $2 + 0$ and 1^+ as a function of the total $^0P_{12}(2)$ pump power. The depletion ratio decreases as the pump power increases, indicating more molecules were pumped from $N = 1$. However, the diminishing gradient of this trend with higher pump powers suggests either that the optical pumping is not perfect or that the extra, unidentified lines in the R(1) spectrum contribute an irreducible background in this measurement. Without additional data, it is challenging to predict if this trend would continue to $\bar{R} = 0$ at higher pump powers. The results presented in section 6.3.2 reveal that at these powers (~ 22 mW per sideband), there is inadequate power to completely deplete the $X^2\Sigma^+$ ($N = 2$) states, which also implies insufficient power to fully deplete the $X^2\Sigma^+$ ($N = 1$) states.

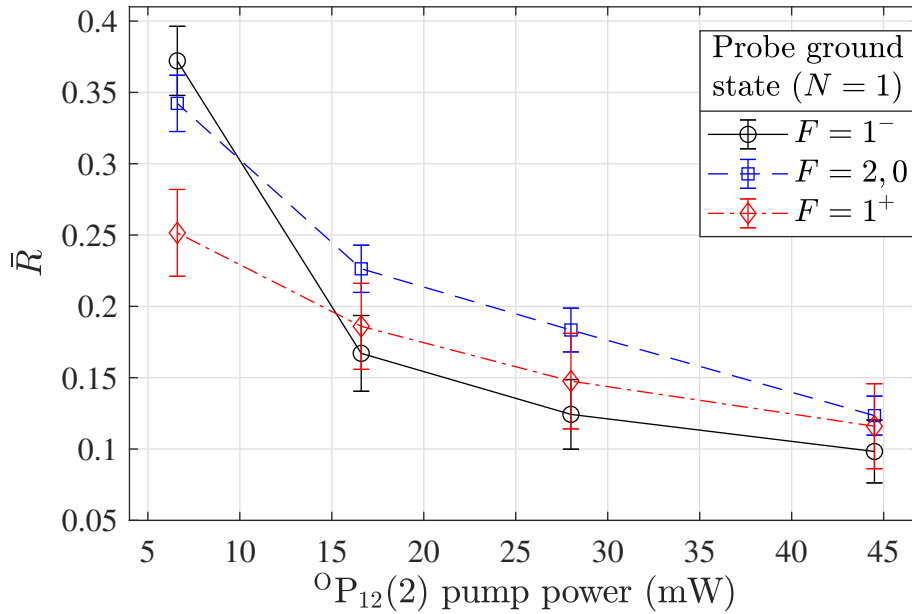


Figure 6.7: Depletion ratio of the mean LIF signals obtained with and without pumping, measured using the R(1) transitions for probing, at different total $^0P_{12}(2)$ pump laser powers.

Dependency on microwave frequency

This experiment aimed to investigate the effect of microwave frequency on the pumping efficiency of $X^2\Sigma^+$ ($N = 1$) hyperfine states by varying the frequency of the oscillator used for creating the mi-

crowave sidebands. Since the microwave mixing scheme used off-resonant pumping, it was important to confirm that the pumping was effectively addressing the desired microwave transitions.

Figure 6.8 displays the frequencies of the applied microwave fields alongside the transition resonances, with the frequency zero-point at the centre frequency (28.9507 GHz) of the microwave sidebands used for other measurements in this chapter. In section 3.4, I describe how a 15.6 MHz waveform was mixed with a microwave waveform to create the two microwave sidebands needed for addressing the $F_1 - F_4$ transitions. By varying the frequency of the waveform outputted by the microwave synthesiser between -2 MHz and 2 MHz, both sideband frequencies were shifted by -4 MHz to 4 MHz; note that the synthesiser frequency was doubled before the sidebands were created. The microwave power used in this experiment was kept low to minimise power-broadening effects on the transitions, with only 14.2 dBm being applied at the horn input.

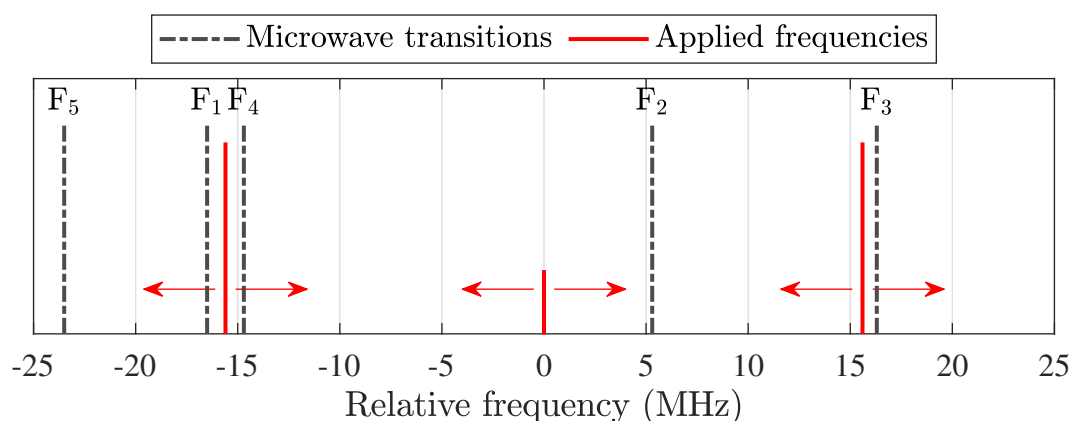


Figure 6.8: Diagram illustrating the frequency shifts used for investigating the dependency of $\bar{R}$ on the applied microwave frequencies. The black dashed lines indicate the transition resonances, while the solid red lines represent the applied microwave sidebands and carrier frequency, centred at 0 MHz. The carrier frequency and, in turn, the two microwave sidebands were shifted through the range ± 4 MHz as indicated by the horizontal arrows.

The $Q(0)$ pump laser was not used in this experiment to avoid decreasing the photon scattering rate, which could affect the pumping efficiency of molecules out of $X^2\Sigma^+(N=1)$. However, a 44.2 mW $^{10}\text{P}_{12}(2)$ pump laser was used, where the power was split evenly between its sidebands. A single-frequency, 1.1 mW, probe laser was used to address the $X^2\Sigma^+(N=1)$ hyperfine states. Three separate measurements were performed, one for each of the $R(1)$ peaks shown in figure 6.6, and with the microwave centre frequency being scanned over the same range.

Figure 6.9 shows the mean depletion ratio, $\bar{R}$, as a function of relative frequency, with data sets for each of the $X^2\Sigma^+$ ($N = 1$) hyperfine states being addressed by the probe light. The curve, $g(f)$, was fitted to each data set:

$$g(f) = a - b \frac{\gamma}{\pi(\gamma^2 + (f - f_0)^2)} \quad (6.1)$$

where f was the frequency. The first term, a , represents a baseline, which was expected to be equal to one because it represents the ratio of pumped/unpumped molecules far from microwave resonance. The second term was a negative Lorentzian, with width, γ , and centre frequency, f_0 , multiplied by a factor, b .

In figure 6.9, results for the $F = 1^-$ and $F = 1^+$ hyperfine states are shown by the black circles and red diamonds, respectively. The baseline for each fit was $a = 1.07(5)$ and $a = 1.01(19)$, where the numbers in parentheses represent the standard error calculated using the 95% confidence interval of the fit. These values agree with the expected value of $a = 1$. However, the baseline for the $F = 1^+$ fit has a higher uncertainty due to the decrease in molecular flux between measurements, leading to increased statistical noise.

The second term of the $F = 1^-$ fit was centered at $f_0 = -0.93(5)$ MHz with a width of $\gamma = 2.00(16)$ MHz, while the $F = 1^+$ fit was centered at $f_0 = 0.46(16)$ MHz with $\gamma = 2.6(7)$ MHz. The expected centre frequencies for the $F = 1^-$ and $F = 1^+$ transitions, calculated using spectroscopic data from Lim et al. (2017) [56], were $f_0 = -1.0(2)$ MHz and $f_0 = 0.8(2)$ MHz, respectively. The centre frequency for the $F = 1^-$ fit was consistent with the expected value. The agreement between the expected value and the centre frequency of the $F = 1^+$ fit was not as good, but the two values were not significantly different at the 90% confidence level.

The crossing point of the two fits³ is at $f = -0.3$ MHz, indicating the point at which both transitions are pumped with equal efficiency. To optimally address the two transitions in this low microwave power regime, the sideband frequency addressing them would need to be decreased by

³Multiple roots exist for the two fits, but only the root between the minimums of the two fits is considered here.

this amount. However, data taken with microwaves at maximum power demonstrated significant power broadening that negates this inefficiency.

The other microwave sideband addressed the F_2 and F_3 transitions. In figure 6.9, data for the $F = 2$ and $F = 0$ states are presented, represented by blue squares. They follow the same trend as the other curves, and the $g(f)$ fit achieved an adjusted R-square of 0.99. The fit's baseline was $a = 1.012(37)$, consistent with the expected value of $a = 1$. The fit resulted in a centre frequency of $f_0 = 0.60(5)$ MHz and a full width at half maximum of $\gamma = 1.67(14)$ MHz. At the 90% confidence level, the centre frequency agreed with the expected value of $f_0 = 0.4(2)$ MHz.

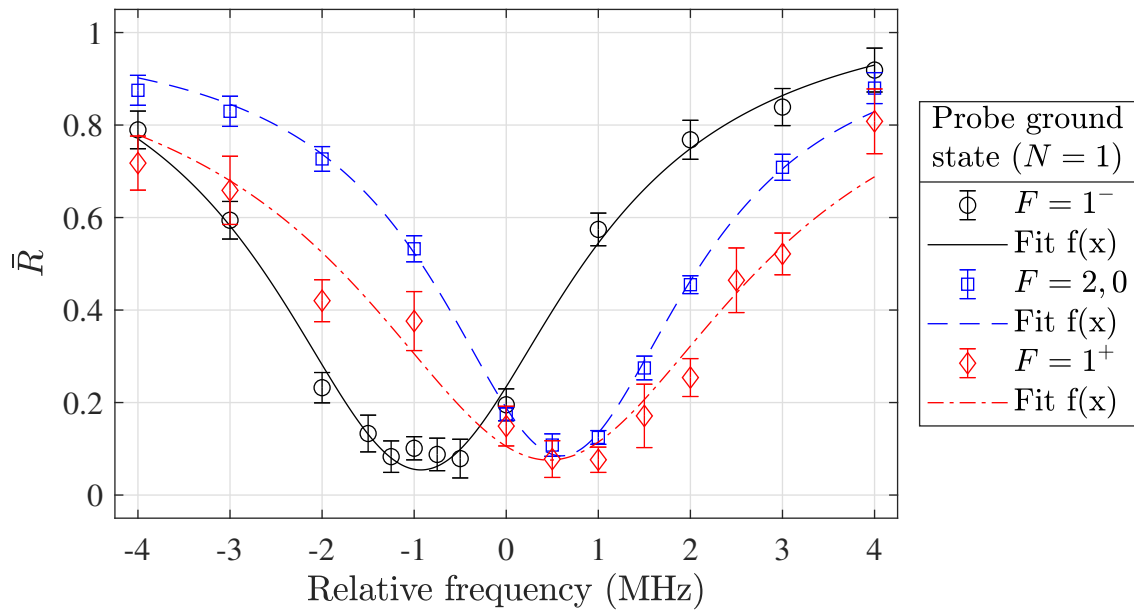


Figure 6.9: Microwave depletion spectroscopy of the $X^2\Sigma^+$ ($N = 1$) hyperfine states, showing the mean depletion ratio $\bar{R}$ as a function of the relative microwave frequency. The black circles and red diamonds represent the $F = 1^-$ and $F = 1^+$ hyperfine states, respectively, while the blue squares represent the $F = 2$ and $F = 0$ hyperfine states, which are indistinguishable. The lines are fits of the function $g(f)$ to the data sets.

Dependency on microwave power

The goal of this study was to examine the impact of microwave power on the pumping efficiency of hyperfine states $X^2\Sigma^+$ ($N = 1$) by varying the power output of the microwave synthesiser. As the microwave mixing scheme used off-resonant pumping, high-power microwaves were required. Therefore, confirming that the transitions of interest were being efficiently addressed was essential.

For optical pumping of the molecules, both pump lasers were used, namely 11 mW of Q(0) light and 44.2 mW of $^{\circ}\text{P}_{12}(2)$ light. The microwave power input at the horn was adjusted between 5.2 dBm and 24.2 dBm per sideband. To measure the population in the $X^2\Sigma^+(N = 1)$ hyperfine states, a probe laser with a single frequency of 1.5 mW was utilised. Three separate measurements were performed, one for each of the R(1) peaks shown in figure 6.6, with the microwave power scanned over the same range in each measurement.

Figure 6.10 shows the depletion ratio of the $X^2\Sigma^+(N = 1)$ hyperfine states $F = 1^-$, 2 and 1^+ as a function of microwave power. As expected, the depletion ratio decreases with increasing microwave power, indicating more molecules were pumped from $N = 1$. However, this trend shows rapidly diminishing returns at higher microwave powers. Therefore, it's unlikely that increasing microwave power beyond the range shown here would produce significant improvements. Notably, the data show a similar trend to when the power of the $^{\circ}\text{P}_{12}(2)$ pump laser was varied (see figure 6.7), which is unsurprising given that both parameters play a similar role in the pumping scheme.

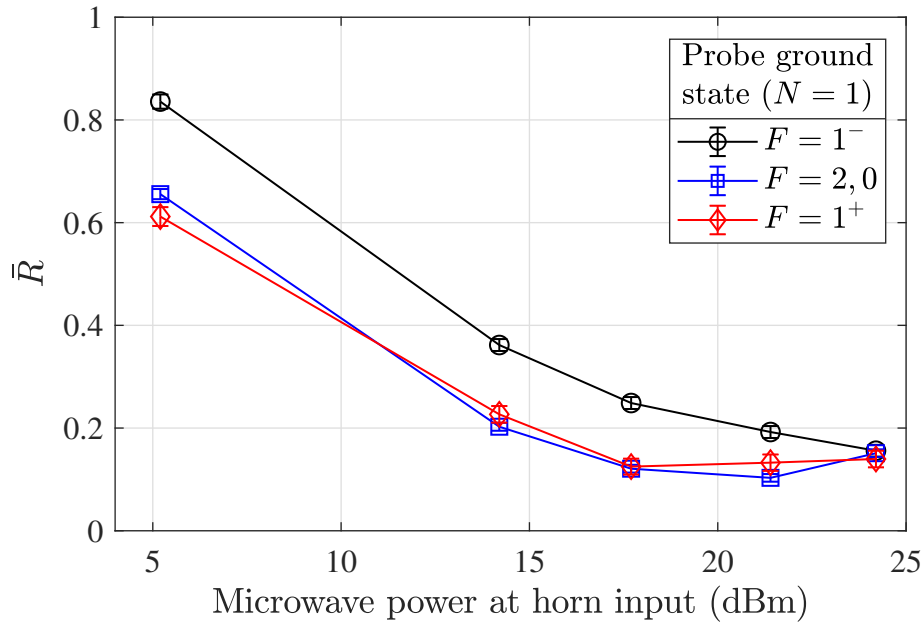


Figure 6.10: Depletion ratio, $\bar{R}$, of the $X^2\Sigma^+(N = 1)$ hyperfine states as a function of microwave power input. The black circles, blue squares and red diamonds represent the $F = 1^-$, $2 + 0$ and $F = 1^+$ hyperfine states, respectively.

6.3.2 Depletion of the $N=2$ hyperfine states

This section focuses on the factors affecting the efficiency of pumping molecules from the $X^2\Sigma^+$ ($N = 2$) rotational state. The pumping scheme's effectiveness depends on various factors, including the power of the pump light and microwave. In a power-limited experiment, adding more transitions to the pumping scheme can reduce the scattering rate and, consequently, decrease the pumping efficiency.

Depletion ratio measurements were conducted for each distinguishable set of transitions in the $^{\circ}\text{P}_{12}(2)$ branch used to probe the $X^2\Sigma^+$ ($N = 2$) population. A single-frequency laser with power 1.9 mW was used to measure the populations in the $X^2\Sigma^+$ ($N = 2$) hyperfine states. For each measurement set, the $^{\circ}\text{P}_{12}(2)$ pump light was always used, and four measurements were taken by varying the application of other pumping transitions: no microwave or Q(0) light, microwaves only, Q(0) only, and both microwaves and Q(0). In each measurement, the $^{\circ}\text{P}_{12}(2)$ pump power was varied. The 0 MHz sideband was scanned from 1 mW to 73 mW, and the 185 MHz sideband scanned from 0.4 mW to 34 mW. The Q(0) laser had a power of 36.5 mW when used, and the microwaves had a maximum power of 24.2 dBm when applied.

The results of the experiment are presented in figure 6.11. The data sets exhibit a consistent trend, where the depletion ratio decreases rapidly with increasing $^{\circ}\text{P}_{12}(2)$ pump power and then plateaus, with further power increases resulting in diminishing returns. As anticipated, using microwaves or Q(0) pump light resulted in a lower scattering rate and more population pumped towards $N = 2$, leading to less depletion of the $X^2\Sigma^+$ ($N = 2$) hyperfine states.

Figure 6.11a shows the depletion ratio measurements for the $X^2\Sigma^+$ ($N = 2$, $F = 1, 2^+$) states. The results indicate that the introduction of Q(0) pump light led to a slight decrease in the scattering rate, resulting in a slightly slower decrease of $\bar{R}$ with increasing $^{\circ}\text{P}_{12}(2)$ pump power. However, when microwaves were added to the pumping scheme, the scattering rate was significantly reduced, and the $\bar{R}$ plateau increased. This marked difference is likely attributable to the microwaves adding three extra states from $N = 1$ to the pumping scheme, greatly reducing the scattering rate and coupling all of the $N = 1$ population to $N = 2$.

Figure 6.11b shows the depletion ratio measurements for the $X^2\Sigma^+ (N = 2, F = 2^-)$ state. The trends are similar to those in figure 6.11a, except that adding microwaves alone did not significantly reduce the scattering rate. This was likely because only $X^2\Sigma^+ (N = 1, F = 1^-)$ was connected to $X^2\Sigma^+ (N = 2, F = 2^-)$ using microwaves.

The results presented in figure 6.11 are promising; however, it is currently unclear which experiment parameters must be adjusted to improve pumping efficiency when microwaves and Q(0) pump light are applied. To better understand the limitations, further investigation into the duration of molecular interaction with the pump light could be valuable.

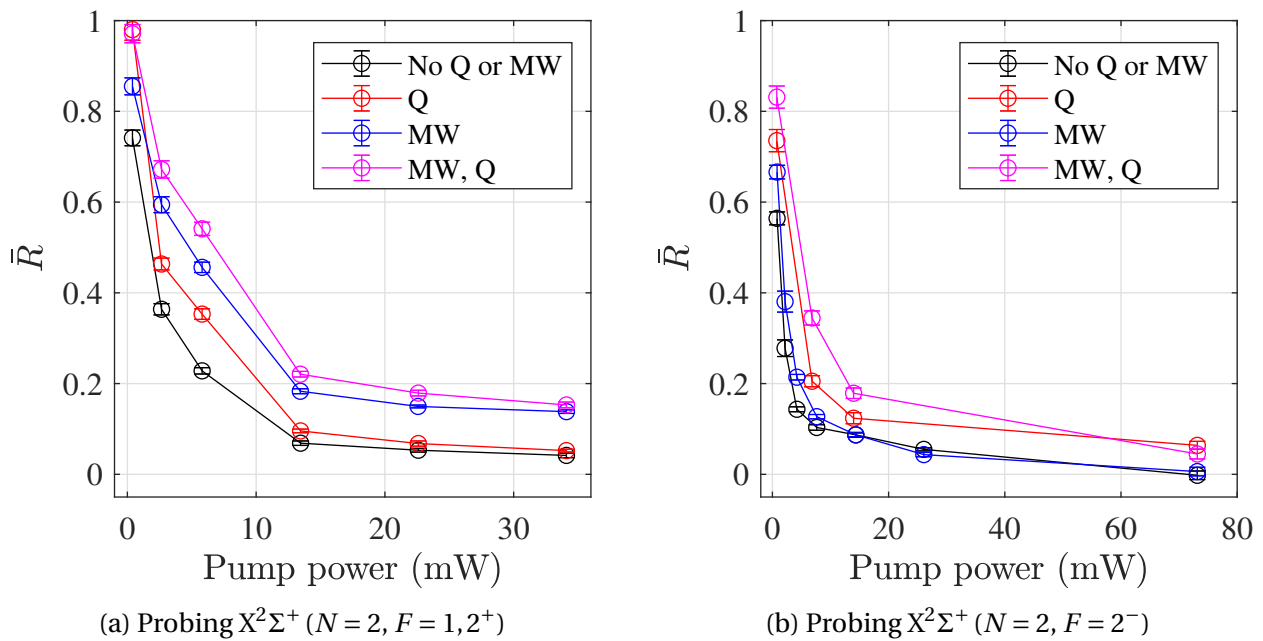


Figure 6.11: Depletion ratio measurements for the $X^2\Sigma^+ (N = 2)$ hyperfine states $F = 2^-$, 1 and 2^+ . Each plot shows $\bar{R}$ as a function of the sideband pump power applied to the state(s) being probed. The legends highlight when microwaves (MW) and Q(0) pumping light were applied.

6.3.3 Accumulation and depletion of the N=0 hyperfine states

This section presents measurements of the accumulation and depletion of molecules in the two $X^2\Sigma^+ (N = 0, F = 0, 1)$ hyperfine states. For these measurements, a single-frequency 1.5 mW probe laser was scanned in frequency over the Q(0) transitions whilst pumping was applied to the molecules. It was expected that the $X^2\Sigma^+ (N = 0, F = 1)$ state would become depleted since it was being addressed by pumping light, while the $X^2\Sigma^+ (N = 0, F = 0)$ state would accrete not only molecules

from the $X^2\Sigma^+$ ($N = 0$, $F = 1$) state, but also those from the higher rotational states, $N = 1$ and $N = 2$. These measurements were performed early in optimising the pumping scheme, so whilst they do not reflect the final pumping efficiency, they approximately demonstrate the effect of pumping on the $N = 0$ hyperfine states.

The microwave power was held constant at full power (24.2 dBm per sideband) during the measurements, but the sideband addressing the F_2 and F_3 transitions was not optimally tuned for pumping the F_3 transition. The molecules were subjected to 30 mW of Q(0) pump light in all of the measurements. To investigate the effect of the $^{\text{O}}\text{P}_{12}(2)$ pump power on the accumulation of the $X^2\Sigma^+$ ($N = 0$, $F = 0$) state, three measurements were performed with pump powers of 8, 34 and 120 mW. However, it was later discovered that the unshifted sideband of the $^{\text{O}}\text{P}_{12}(2)$ laser had poor fibre coupling, resulting in approximately 90% of the power being derived from the 185 MHz sideband.

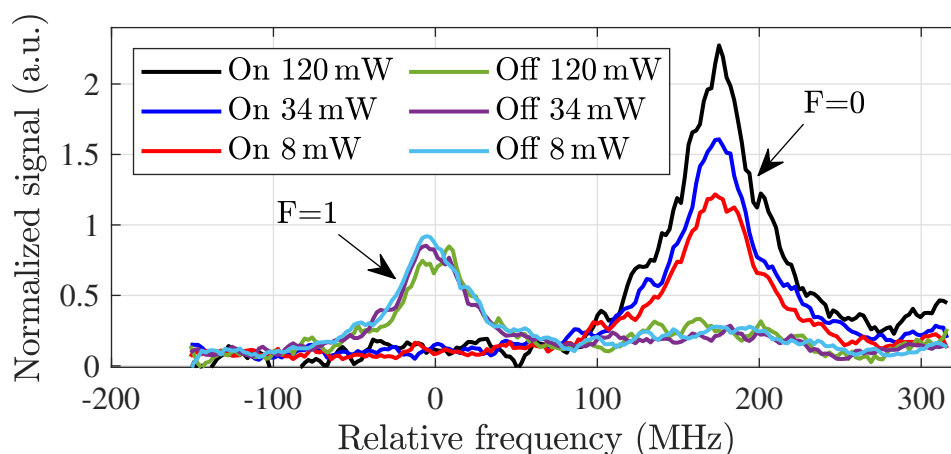


Figure 6.12: Normalized laser-induced fluorescence (LIF) measurements of the $X^2\Sigma^+$ ($N = 0$) hyperfine states as a function of the probe laser frequency. Three different $^{\text{O}}\text{P}_{12}(2)$ pump powers were used: 8, 34, and 120 mW, with the pumping turned on and off for each power. The LIF signals were normalised to the height of the $F = 1$ peak to account for variations in the molecule flux from the source between measurements.

The measurements for the three $^{\text{O}}\text{P}_{12}(2)$ pump powers explored are presented in Figure 6.12. Two curves are plotted for each power, one with the pumping turned on, and one with it turned off. To account for the variation in molecule flux produced by the source between measurements, each pair of curves is normalised to the height of the $F = 1$ peak with the optical pumping turned off. At low pump power, the $F = 0$ peak is similar in size to the $F = 1$ peak, indicating the potential

transfer of molecules between the $N = 0$ hyperfine states. However, as the pump power increases, the $F = 0$ LIF signal continues to increase after the $F = 1$ state is fully depleted, suggesting that molecules from other states are accreting in the target state of the pumping scheme. Although not performed under optimal conditions, these results demonstrate the effectiveness of the pumping scheme.

6.4 Summary

In this chapter, a new pumping scheme is presented, which has been developed to transfer YbF molecules from the $X^2\Sigma^+$ ($N = 1$) rotational state to the $X^2\Sigma^+$ ($N = 0, F = 0$) hyperfine state for the purposes of an eEDM measurement. The scheme used microwaves to connect the $X^2\Sigma^+$ ($N = 1$) and ($N = 2$) rotational states and laser light to pump molecules into the target state. The depletion ratio of the $N = 1$ state could be driven to $0.1 - 0.15$ with sufficient pump laser and microwave power, with potential improvement if $^{\text{O}}\text{P}_{12}(2)$ pump power is increased. Similar depletion ratios were demonstrated for the $N = 2$ states, but further investigation is needed into the effect of pumping interaction time. Results showing the accretion and depletion of molecules in the $N = 0$ hyperfine states demonstrate that the scheme transfers molecules to the intended target state. Further investigation into the pumping interaction time and depletion of the $N = 2$ states is suggested to improve efficiency. Once equipment repairs are completed, the F_5 microwave transition should be addressed to investigate hypothesised dark states. Overall, the new pumping scheme has much potential for eEDM measurements.

Chapter 7

Conclusions and outlook

7.1 Enhancements and additions to the experiment

The new YbF electron EDM experiment comprises multiple components. I have contributed to several aspects preceding the electron EDM measurement region, which has recently been completed. The scope of my work has encompassed a diverse range of accomplishments, including the development of a new YbF molecular source, the implementation of two-dimensional laser cooling techniques for molecular beam collimation, the design and assembly of a magnetic lens to facilitate molecular beam focusing, and the development of a new pumping scheme to prepare the molecules for the electron EDM measurement. In the following section, I provide a concise overview of my achievements in each of these domains.

7.1.1 The YbF molecular source

The experiment commenced with the production of a YbF molecule beam. Although not the central focus of this thesis, we dedicated efforts to building and optimising the new YbF buffer gas-cooled source based on the design outlined by Hutzler et al. (2012) [43]. A comprehensive description of our source can be found in chapter 3, complete with detailed component drawings. This upgrade aimed to increase molecular flux significantly; however, after thoroughly exploring the

experimental parameter space, we observed only marginal improvements compared to our previous design. Nevertheless, the new apparatus boasted enhanced practicality, featuring a custom design incorporating valuable knowledge accumulated from experiments conducted at Imperial. Notably, the vacuum chamber door stood out as a personal favourite, providing convenient access during optimisation processes that required the frequent opening of the vacuum chamber.

Accompanying this upgrade, I developed custom-designed software (detailed in section 3.5.3) to control various aspects of the experiment by integrating different experimental hardware with the main lab computer. The software was a central hub for monitoring pressure and temperature throughout the setup. It automated the controlled heating and cooling of the source, allowing us to focus on other critical tasks. The software remains a pivotal component of the experiment and has been expanded upon by others to incorporate control of experimental hardware in the EDM measurement region.

7.1.2 Two-dimensional laser cooling of YbF

Undoubtedly, a significant accomplishment of this work was the development and comprehensive characterisation of a two-dimensional laser cooling scheme for the YbF molecular beam, which is the topic of chapter 4. Sub-Doppler Sisyphus laser cooling was successfully demonstrated for both transverse axes, initially exploring one-dimensional cooling in each axis and later addressing both axes simultaneously to collimate the molecular beam. After upgrading the cooling laser, the challenge of handling the hyperfine structure in a large detuning configuration was addressed, as outlined in section 4.3.1. The investigation identified an optimal configuration where the sideband addressing the $X^2\Sigma^+(\nu = 0, N = 1, F = 1^-)$ hyperfine state was entirely eliminated. This observation was attributed to the high-intensity configuration, where the large detuning required for cooling caused this sideband to become near red-detuned from the $F = 2, 0$ states, resulting in competing heating effects that countered the blue-detuned cooling of other hyperfine states.

The results of the two-dimensional laser cooling experiments are presented in section 4.4. The dependence of magnetic field-induced laser cooling on the applied magnetic field was demon-

strated, with the expected minimum achieved at zero field. At applied field strengths above 0.5 G, this method yielded a higher population of ultracold molecules than polarisation gradient cooling. Furthermore, the investigations into the relationship between laser cooling and repump laser powers revealed that although increasing cooling laser power could produce more ultracold molecules, the available repump laser power was sufficient to efficiently repump the molecules, even with the increased cooling light power.

In section 4.4.4, characterisation of the transverse temperature of the laser-cooled molecular beam is presented, considering variables such as cooling light detuning, applied magnetic field strength, and helium flow in the molecular source. The measurements consistently approached the resolution limit, indicating that the transverse temperature was below 200 μK under optimal experimental conditions. Moreover, estimations outlined in section 4.4.5 demonstrated that the beam's brightness increased by more than two orders of magnitude when laser cooling was implemented.

7.1.3 Molecular funnel

Chapter 5 details the development of a magnetic lens designed to guide the molecular beam to the laser-cooling region, increasing the number of laser-cooled molecules. While the concept of combining a magnetic lens with laser cooling has been successfully demonstrated with atomic beams, known as an atomic funnel, the realisation of a molecular funnel has remained a challenge, primarily due to the complexities of laser cooling molecules. However, this is no longer a limitation for YbF. Simulations of a magnetic lens were conducted using the methods described in section 5.2, typically employed for high-energy particle beam systems but successfully adapted for this purpose. The simulations aimed to determine optimal magnetic lens parameters that would overlap the molecular phase space distribution with the laser cooling phase space acceptance, maximising the number of molecules effectively subjected to laser cooling.

The results of the simulations, presented in section 5.3, demonstrate that the chosen magnetic lens design efficiently guides molecules over a wide range of longitudinal velocities, approximately $\Delta v_z \approx 50, \text{ms}^{-1}$. By adjusting the longitudinal position of the lens, the efficiency of guiding molecules

can be tailored, with closer positioning to the source favouring more efficient guidance of slower molecules and vice versa. The simulations indicate a potential increase in the brightness of the laser-cooled molecular beam by a factor of 5.3. This substantial enhancement, combined with the collimation achieved through laser cooling, can potentially improve the sensitivity of electron EDM measurements by a factor of 2.3.

Based on the simulation results, a suitable design for the magnetic lens was chosen and is outlined in section 5.4, along with comprehensive information regarding its assembly and characterisation. The magnetic lens has been fully assembled and is currently awaiting installation in the beamline, marking a significant step toward realising a molecular funnel.

7.1.4 Preparing the cooled molecules for the eEDM measurement

Chapter 6 focuses on the crucial step of transferring laser-cooled YbF molecules to the absolute ground state, a prerequisite for the electron EDM measurement process. Initially, the laser-cooled molecules reside in the first excited rotational state, $N = 1$. However, the electron EDM measurement process necessitates their initial preparation in the absolute ground state, $X^2\Sigma^+(\nu = 0, N = 0, F = 0)$. To address this, a new scheme has been developed to facilitate the transfer from the laser cooling state to the absolute ground state.

The newly developed scheme, described in section 6.1, utilises microwaves to establish connections between the first and second excited rotational states. Optical pumping is then employed to transfer molecules from the $(N = 2)$ and $(N = 0, F = 1)$ states. The results, presented in section 6.3, assess various aspects of the pumping scheme, including depletion ratios for the rotational states $N = 0, 1$, and 2 under different experimental conditions. It is important to note that one of the microwave frequencies required to address a dark state was not included in these measurements due to an equipment malfunction. Nonetheless, the obtained results were highly promising, exhibiting depletion ratios of 0.1–0.15 for the $N = 1$ hyperfine states and even more favourable depletion ratios for the $N = 2$ hyperfine states.

Measurements of the ground rotational state revealed two significant findings. Firstly, the popu-

lation in the ($N = 0, F = 1$) state was nearly completely depleted through optical pumping. Secondly, a notable accumulation of molecules was observed in the ($N = 0, F = 0$) state, indicating a substantial increase in population within the target state. Ongoing optimisation efforts continue in this area; however, the results thus far are highly encouraging, affirming the efficacy of the new pumping scheme in preparing the molecules for electron EDM measurements.

7.2 Sensitivity of an eEDM measurement

In section 4.4.6, I discussed the potential sensitivity of an electron EDM measurement using our two-dimensionally laser-cooled YbF beam. The estimation was based on real-world parameters obtained from the YbF apparatus used in the 2011 measurement, which has undergone continuous development over the years [2, 3, 41, 100]. After one day of measurement, we anticipate a statistical uncertainty of $\sigma_{d_e}^{(\text{laser cooling})} = 5 \times 10^{-30} \text{ e cm}$. This value is projected to increase by an order of magnitude after one hundred days:

$$\sigma_{d_e}^{(\text{laser cooling})} = 5 \times 10^{-31} \text{ e cm.} \quad (7.1)$$

Assuming we adequately account for systematic sources of measurement noise, this level of uncertainty would be close to the recent achievement of the Cornell Group, which reported $4.1 \times 10^{-30} \text{ e cm}$ [16]. However, simulations suggest that incorporating the magnetic lens, which creates a molecular funnel, will enhance beam brightness more than five-fold and further improve the prediction stated in equation 7.1. The expected improvement of a factor of 2.3 would yield,

$$\sigma_{d_e}^{(\text{mol. funnel})} = 2.2 \times 10^{-31} \text{ e cm.} \quad (7.2)$$

Achieving such a measurement would be world-leading, surpassing the statistical uncertainty of the Cornell Group by approximately more than an order of magnitude.

7.3 Conclusions

In conclusion, this thesis marks a significant breakthrough in our ability to control the YbF molecular beam. Our newly developed molecular source yields a slow, cold beam that undergoes two-dimensional laser cooling in the transverse axes, resulting in effective collimation. This achievement holds tremendous potential for conducting an exceptionally precise electron EDM measurement, thanks to the extended interaction time and enhanced molecular coherence.

Moreover, this work serves as a foundation for further advancements. With the magnetic lens poised for installation, we anticipate a substantial boost in measurement sensitivity, potentially reaching world-leading levels of precision. In conjunction with a novel pumping scheme designed to prepare the molecules specifically for the electron EDM measurement, our experimental setup is rapidly approaching the point where we can embark on actual electron EDM measurements.

The author eagerly awaits the publication of future results in the scientific literature, as this groundbreaking apparatus holds immense promise for advancing our understanding of electron EDM and pushing the boundaries of precision measurement.

7.4 Outlook

The outlook for the experiment is highly promising. The groundwork laid by this work paves the way for the commencement of electron EDM measurements. While there is still some work ahead to unravel the reasons behind the incomplete transfer of molecules from the laser cooling state to the absolute ground state, the current scheme is already exhibiting excellent performance.

Dedicated efforts from several members of our group have been focused on designing STIRAP techniques, and they are currently in the process of implementing these techniques in the experimental setup. The EDM interaction region has been meticulously constructed and thoroughly characterised to identify and eliminate systematic magnetic noise sources. Additionally, a degaussing setup has been implemented to eradicate remnant magnetic fields present in the magnetic shields surrounding the interaction region.

Others have undertaken extensive work characterising the electric field plates and mitigating leakage currents. The interaction and detection regions have been fully assembled and seamlessly integrated into the overall beamline configuration. I hope that in the near future, they will be able to initiate the measurement of interference fringes and embark on a comprehensive characterisation of systematic noise sources using the molecular beam itself.

This outlook is certainly not exhaustive, and it is essential to acknowledge the significant effort that lies ahead to commence electron EDM measurements. Nonetheless, with the experiment's well-positioned status and progress made thus far, I am optimistic that the electron EDM measurements will soon be underway.

Appendix A

Appendices

A.1 Phase space coordinate transformation

It is common to describe a phase space in terms of velocity, v_w , and position, w . Figure A.1 shows two examples of velocity-position phase space ellipses. The velocity and position of a particle are easily read from the plot.

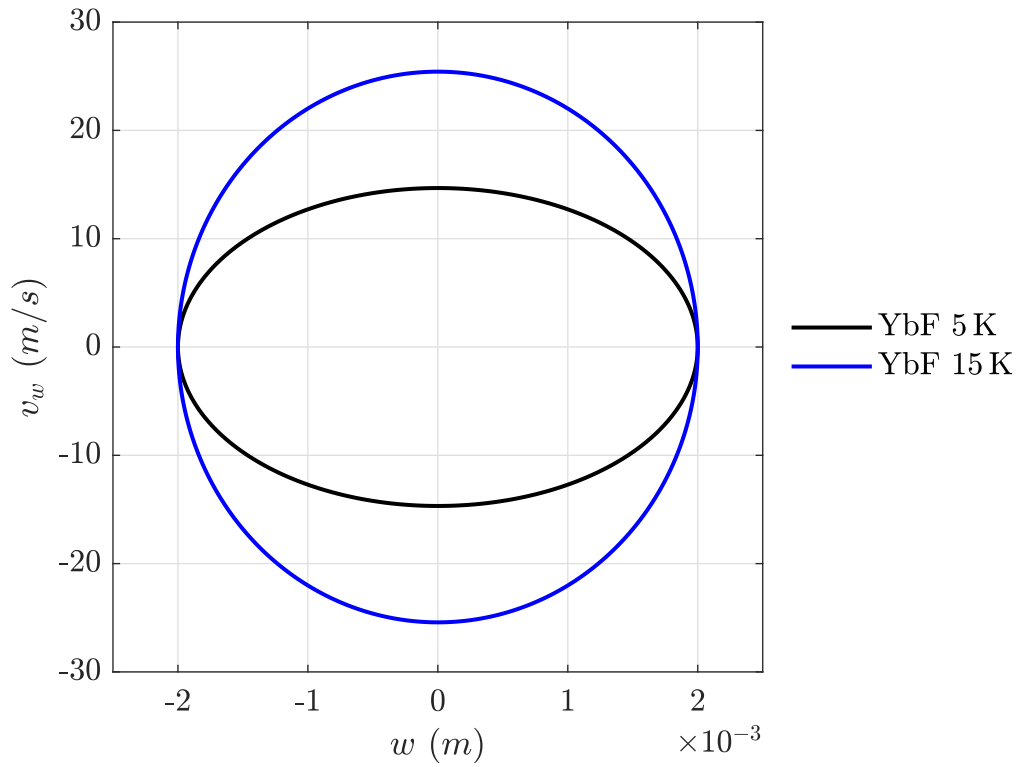


Figure A.1: Simulated phase space ellipses in velocity-position space. The black contour shows a 5 K YbF PSD ellipse whilst the larger blue contour shows a 15 K YbF PSD ellipse for comparison.

However, in this work, the velocity-position phase space was mapped onto another, such that the two axes have the same dimension. Then direct comparisons to the general ellipse equation can be made without running into problems with dimensional analysis.

Consider our velocity-position phase space ellipse, $\epsilon = Aw^2 + 2Bwv_w + Cv_w^2$. To map the velocity axis to one with units of position, we must divide by a frequency, ω , $v_w \mapsto v_w/\omega$. The second moments of the distributions are changed as follows:

$$\langle v_w^2 \rangle \mapsto \frac{\langle v_w^2 \rangle}{\omega^2} = \langle w'^2 \rangle \quad (\text{A.1})$$

$$\langle wv_w \rangle \mapsto \frac{\langle wv_w \rangle}{\omega} = \langle ww' \rangle \quad (\text{A.2})$$

$$\langle w^2 \rangle \mapsto \langle w^2 \rangle \quad (\text{A.3})$$

Substitution into equation 5.30 then shows that the emittance, ϵ , must be altered by a factor $1/\omega$.

Then using equations 5.20-5.22, we find the following transformation for the ellipse parameters:

$$\begin{bmatrix} A \\ B \\ C \end{bmatrix} \mapsto \begin{bmatrix} \frac{1}{\omega} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \omega \end{bmatrix} \cdot \begin{bmatrix} A \\ B \\ C \end{bmatrix} = \begin{bmatrix} \gamma \\ \alpha \\ \beta \end{bmatrix} \quad (\text{A.4})$$

where the final vector is formed of the Twiss parameters outlined in section 5.2. Note how the transformation is consistent with the identity, $\gamma\beta - \alpha^2 = 1$.

Applying this mapping to the ellipses in figure A.1, using the lens frequency from section 5.3, results in the phase space ellipses shown in figure A.2.

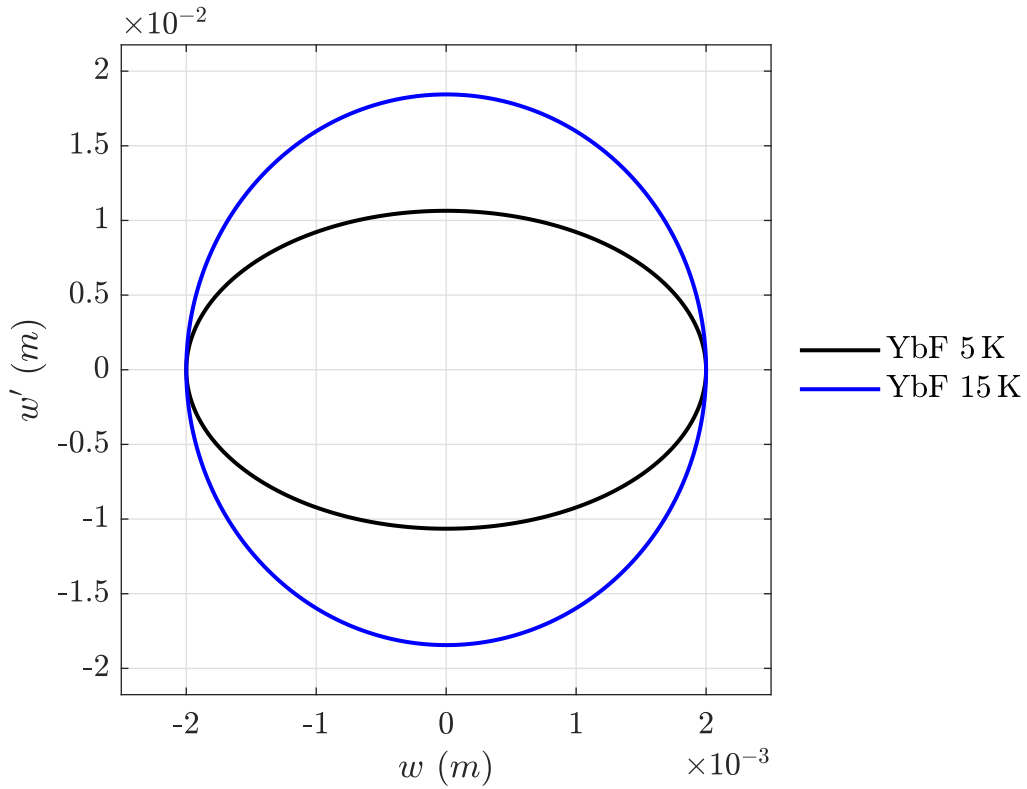


Figure A.2: Simulation of phase space ellipses mapped from velocity-position space to position-position space using lens frequency $\omega = 1.38 \text{ krad s}^{-1}$. The black contour shows a 5 K YbF PSD ellipse, whilst the larger blue contour shows a 15 K YbF PSD ellipse for comparison.

If the frequency of the magnetic lens is used for ω , then the ellipse simply rotates as it evolves through the lens.

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