

Observation of ‘Missing’ Vibrational Levels and Extending Photoassociation Spectra of Excited Ultracold Cesium Dimers

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A thesis submitted to
Institute of Experimental Physics
Faculty of Physics of the University of Warsaw
in partial fulfillment of the requirements
for the degree of Doctor of Philosophy

March 2024

Abstract

This thesis reports on photoassociation spectroscopy experiments on ultracold cesium molecules, achieved through a series of benchmark measurements conducted with the apparatus originally designed for studies of ultracold KCs molecules. The development of the experimental setup, its characterization and testing are described in details, with an emphasis on the methods required to enhance the efficiency of the photoassociation process. A laser system consisting of two commercial, narrow-linewidth titanium sapphire lasers phase-locked to an optical frequency comb has been constructed to facilitate future single- and two-color photoassociation spectroscopy of Cs_2 , K_2 and KCs molecules. The hardware development has been crucial to the results described in this thesis and will be essential for the ongoing work to produce ultracold KCs molecules in their rotational and vibrational ground states.

The main result of the thesis is the detection of two ‘missing’, lowest-lying vibrational levels in the outer-well of the $0_g^-(6S_{1/2} + 6P_{3/2})$ molecular potential. According to theoretical predictions of Bouloufa et al. (Physical Review A, 75(5):052501, 2007), the introduction of these levels is necessary to provide a numerical form of the molecular potential curve that would explain existing experimental data available for the states located in this long-range well. The measurements performed as a part of this work have provided an unambiguous confirmation of theoretical predictions, thus concluding several unsuccessful attempts made by other research groups. With this work, a proof is provided that a previous study that claimed the discovery of these ‘missing’ levels in reality reported levels belonging to another molecular potential. As a complementary investigation, the same experimental apparatus was utilized to extend the observed range of vibrational energy levels of other molecular states $0_u^+(6S_{1/2} + 6P_{3/2})$ and $1_g(6S_{1/2} + 6P_{3/2})$ to -300 cm^{-1} from the $(6S_{1/2} + 6P_{3/2})$ asymptote, surpassing the previously reported experimental limit of -120 cm^{-1} .

Following these observations, the potential energy curves of the molecular states under study were numerically reviewed. The analysis of states $0_u^+(6S_{1/2} + 6P_{3/2})$ and $1_g(6S_{1/2} + 6P_{3/2})$ indicates that the new measurements cannot be sufficiently well described by molecular models used for long-range interactions, necessitating the development of a refined model formulated for the intermediate range. However, the numbering of the measured levels seems to be unambiguous. On the other hand, the analysis of the potential energy curve of the $0_g^-(6S_{1/2} + 6P_{3/2})$ state yields results in agreement with the developed theoretical postulates. The relatively small impact of the accurately determined binding energy of the ‘missing’ levels (better than 1 MHz) on the overall shape of the molecular potential is the result of relying on experimental data for numerous vibrational levels that were determined by other research groups with an accuracy of not better than 60 MHz. More precise measurements of at least some of the already-known vibrational

energy levels would be beneficial for the improvement of the potential energy curve and the more accurate determination of important atomic and molecular constants.

The experimental apparatus benchmarked by performing photoassociation spectroscopy of ultracold cesium dimers shows great promise for further studies of ultracold molecules. In the future, it will enable photoassociation spectroscopy of excited molecular states of KCs, aiming to facilitate the formation of ground-state KCs molecules. Additionally, the titanium sapphire laser setup will be essential for ongoing studies of the KAg and CsAg molecules.

Streszczenie

Niniejsza praca demonstruje wyniki spektroskopii fotoasocjacyjnej ultrazimnych cząsteczek cezu, otrzymanych w wyniku serii pomiarów przeprowadzonych przy użyciu układu skonstruowanego do badania ultrazimnych cząsteczek KCs. W pracy szczegółowo opisano rozwój układu eksperymentalnego, jego charakterystyka, ze szczególnym uwzględnieniem metod wymaganych do zwiększenia wydajności procesu fotoasocjacji. Skonstruowano układ laserowy złożony z dwóch komercyjnych, wąskopasmowych laserów tytanowo-szafirowych dowiązanych fazowo do optycznego grzebieńa częstotliwości, co umożliwi w przyszłości spektroskopię jedno- i dwu-fotonową cząsteczek Cs_2 , K_2 i KCs. Rozwój bazy sprzętowej był kluczowy dla otrzymania wyników prezentowanych w niniejszej pracy i będzie nie mniej istotny dla będących w toku badań zmierzających do produkcji ultrazimnych cząsteczek KCs w rotacyjnym i oscylacyjnym stanie podstawowym.

Głównym wynikiem tej pracy jest wykrycie dwóch ‘brakujących’, najniższych położonych poziomów oscylacyjnych w zewnętrznej studni potencjału cząsteczkowego $0_g^-(6S_{1/2} + 6P_{3/2})$. Zgodnie z przewidywaniami teoretycznymi Bouloufa i in. (Physical Review A, 75(5):052501, 2007), wprowadzenie tych dodatkowych poziomów jest konieczne, aby stworzyć numeryczną postać krzywej potencjału cząsteczkowego, która pozwoliłaby uzgodnić wyniki teoretyczne z dostępnymi danymi doświadczalnymi dla tej zewnętrznej studni potencjału. Pomiary przeprowadzone w ramach tej pracy dostarczyły jednoznacznego potwierdzenia przewidywań teoretycznych, tym samym podsumowując zakończone niepowodzeniem próby podjęte przez inne grupy doświadczalne. Praca dostarcza również dowodów, że wyniki poprzednich badań, których autorzy twierdzili, że zaobserwowali te ‘brakujące’ poziomy w rzeczywistości zaobserwowali poziomy należące do innego potencjału molekularnego. Jako uzupełnienie powyższych badań, ten sam układ doświadczalny został wykorzystany do rozszerzenia zakresu zaobserwowanych poziomów oscylacyjnych w potencjałach $0_u^+(6S_{1/2} + 6P_{3/2})$ i $1_g(6S_{1/2} + 6P_{3/2})$ do -300 cm^{-1} poniżej asymptoty $(6S_{1/2} + 6P_{3/2})$, znacznie wykraczając poza dotychczasowy zakres danych doświadczalnych, sięgających do -120 cm^{-1} poniżej tej asymptoty.

Wykorzystując nowo zmierzone poziomy energetyczne, numeryczna forma krzywych potencjałów cząsteczkowych badanych w tej pracy została poddana przeglądowi. Analiza potencjałów $0_u^+(6S_{1/2} + 6P_{3/2})$ i $1_g(6S_{1/2} + 6P_{3/2})$ wskazuje, że nowe pomiary nie mogą być wystarczająco dobrze opisane modelami potencjałów rozwiniętych dla oddziaływań długozasięgowych, tym samym wymuszając rozwój modeli skupiających się na średniozasięgowych częściach krzywych potencjałnych. Niemniej jednak numeracja zmierzonych poziomów oscylacyjnych wydaje się być jednoznaczna. Z drugiej strony, analiza potencjału cząsteczkowego $0_g^-(6S_{1/2} + 6P_{3/2})$ daje wyniki zgodne z dotychczasowymi modelami. W rzeczywistości może to wskazywać na fakt, że wysoka dokładność pomiaru energii wiązania tych ‘brakujących’ poziomów (lepsza niż 1 MHz) ma sto-

sunkowo mały wpływ na całkowity kształt potencjału. Wydaje się to być spowodowane głównie faktem, że w analizie wykorzystano pochodzące od różnych grup badawczych dane eksperymentalne otrzymane z dokładnością nie lepszą niż około 60 MHz. To wskazuje, że bardziej precyzyjne pomiary energii wiązania przynajmniej niektórych już znanych poziomów oscylacyjnych korzystnie wpłynie na poprawienie krzywej potencjału i w efekcie na dokładniejsze wyznaczenie istotnych stałych atomowych i cząsteczkowych.

Układ eksperymentalny, przetestowany poprzez przeprowadzenie spektroskopii fotoasocjacyjnej ultrazimnych dimerów cezu, jest obiecującym narzędziem do badania ultrazimnych cząsteczek. W przyszłości pozwoli on na przeprowadzenie spektroskopii fotoasocjacyjnej wzbudzonych potencjałów cząsteczkowych cząsteczki KCs, otwierając tym samym drogę do wytworzenia tych cząsteczek w rotacyjno-oscylacyjnym i elektronowym stanie podstawowym. Dodatkowo układ złożony z dwóch laserów tytanowo-szafirowych będzie kluczowy w nadchodzących badaniach cząsteczek KAg i CsAg.

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Acknowledgments

First and foremost, I express my deepest gratitude to my esteemed supervisors, Prof. Paweł Kowalczyk and Dr. Mariusz Semczuk, for their invaluable guidance, unwavering support, and immense patience throughout the entirety of my doctoral journey. Their expertise, encouragement, and insightful feedback have been instrumental in shaping this thesis and in my growth as a researcher. I also want to extend special thanks to Dr. Mariusz Semczuk for his supervision of the experiments conducted. Throughout the course of this thesis work, without his patience and belief, these novel observations would not have been realized.

Furthermore, I want to express my sincere gratitude to three outstanding doctoral students - Mateusz Bocheński, Jakub Dobosz, and Paweł Arciszewski, who significantly contributed to my doctoral journey. Their collective expertise has been instrumental in shaping the outcome of this thesis.

I wish to extend my sincere appreciation to Göktuğ, Kerem, Recep, and Savaş. Their presence has been an endless source of entertainment and a great source of support, helping me to keep stress and overwhelming work at bay. Their friendship has been a constant source of joy and comfort throughout my doctoral journey. I would also like to extend my gratitude to Can (Wolkas), Edin (LurCellant) and Bora (Predlak). Our shared experiences have not only been entertaining but have also fostered lasting friendships beyond the virtual realms.

I am profoundly grateful to my family, Hulusi, Gülçin, Berkay, and Hasgül, whose unwavering support and boundless patience have been instrumental in the completion of this thesis. With their encouragement and understanding, this work was possible. Their love and belief in me have been the cornerstone of my journey, and I am deeply thankful for their presence in my life.

I want to extend my heartfelt thanks to Izabela for her unwavering support throughout this journey. Her presence has been simply invaluable. Her encouragement and companionship have made this endeavor all the more fulfilling. Words fall beyond the context.

Chapter 1

Introduction

The advancement in techniques for cooling and trapping of atoms brought significant improvements in metrology [1, 2, 3, 4], with measurements performed with ultracold samples playing a crucial role in determining the validity of theoretical models that predict values of important atomic and molecular constants [5, 6]. For example, photoassociation spectroscopy has been a feasible method to experimentally determine the atomic radiative lifetime of atoms [7, 8] and ground state molecular dissociation energies [9, 10].

Photoassociation spectroscopy, which relies on the formation of short-lived excited state molecules via laser-induced transitions between a state composed of a pair of colliding atoms and a final molecular state, provides a powerful tool for studying molecular properties. This technique becomes even more advantageous when applied to ultracold atomic ensembles, where the precise control of initial conditions allows for enhanced manipulation of the system. In this context, the features observed in photoassociation spectroscopy directly correspond to the properties of initial states of ultracold atoms [11, 12]. Without the ability to control these initial conditions (such as in the case of atomic ensembles at room temperature), one can only determine the average behavior of molecules, as various atoms in different states within the same ensemble will contribute to molecule formation [13]. Furthermore, conducting photoassociation measurements in the ultracold regime provides a unique advantage for precisely determining binding energies, rovibrational energy levels, and interaction potentials with exceptional spectral resolution [14]. This is attributed to the minimal spread in the initial kinetic energies of the colliding atoms within this ultra-low temperature range. Consequently, the narrow range of photon frequencies required to drive the resonant photoassociation process enhances the precision and accuracy of the measurements. In addition, photoassociation emerges as a viable tool that enables the creation of ultracold ground state molecules, addressing challenges associated with the direct cooling of molecules, where the molecular samples have relatively low densities and temperatures [15, 16, 17, 18] compared to ensembles formed relying on photoassociation.

The research detailed in this thesis initially aimed at performing photoassociation spectroscopy of KCs molecules as our research group had already laser-cooled isotopes of potassium (^{39}K , ^{40}K , ^{41}K) together with cesium (^{133}Cs) atoms. As we further refined the cooling techniques for these species, evaluating the overall effectiveness of the newly developed photoassociation setup became essential. We therefore decided to conduct benchmark experiments focusing on the

photoassociation of cesium atoms, as there was a wealth of reference experimental data available, thus making it easier to debug potential issues with the setup. This particular photoassociation setup underwent continuous development, guided by feedback obtained through the spectroscopic data analysis and technical issues encountered throughout the progress. These advancements in the photoassociation apparatus and the improvements made in our ultracold sample preparation stages form the foundation of this thesis. It might be worth pointing out that the work reported in this thesis has been made possible not because of the development of any particular new technique but thanks to a systematic approach to the design and development of the experimental setup. The measurements described in this thesis rely on the combination, in one experimental setup, of many state-of-the-art techniques used in ultracold experiments. As such, the main results presented here are the culmination of many developments in the field of ultracold atoms, benefiting from and building upon over two decades of photoassociation spectroscopy studies of ultracold cesium dimers.

The benchmarking experiments primarily focused on the external well of the 0_g^- long-range state of Cs_2 [19] dissociating to the $6S_{1/2} + 6P_{3/2}$ asymptote. To ensure simplicity and clarity throughout this thesis, we designate molecular states as $0_g^-(P_{3/2})$, where the term within the brackets denotes the atom in its excited state, while the other atom is in its ground state $S_{1/2}$. The long-range molecular interactions require the adequate distancing of two atoms from each other. In such states, the electron cloud exerts negligible influence on the binding energy, contrary to the interactions observed at shorter internuclear distances. This specific behavior of such molecules caught the interest of physicists, as their properties became measurable with high precision, ideally with minimal disturbances from the properties of the atoms they comprise. Notably, the long-range state of Cs_2 , specifically the $0_g^-(P_{3/2})$ state distinguished by its double-well potential, has attracted considerable attention. Its external well has been extensively investigated in numerous papers [20, 21, 22]. However, the experimental data, when analyzed, has revealed inconsistencies regarding both the van der Waals coefficient C_6 , which characterizes the long-range interaction between two ground-state cesium atoms, and the triplet scattering length that parametrizes the elastic collision rate of two cesium atoms. Subsequently, the inconsistencies were addressed by the theoretical work of N. Bouloufa et al. [23], in which it was postulated that there should be two additional vibrational levels that remained unobserved in previous experimental works. Substantial efforts have been made to observe and identify these ‘missing’ vibrational levels to resolve inconsistencies arising from earlier experiments [23]. The pursuit of measuring these levels partially concluded with the published experimental work of Zhang et al. [24]. Subsequently, a year later, an experimental review of these vibrational levels was reported [25]. Experimental data from these two works by Zhang et al. [24] and Ma et al. [25] displayed a significant difference in the energies of the two lowest-lying vibrational levels of the $0_g^-(P_{3/2})$ state. There has been no commentary to date addressing the disparities between the two sets of measurements. Additionally, in these papers, the energies of the two lowest-lying vibrational levels were suspiciously close to the energies of vibrational levels of the $0_u^+(P_{3/2})$ state [25]. This unexpected coincidence has motivated us to verify the disparities in the previous measurements. Moreover, the two lowest-lying vibrational levels of $0_g^-(P_{3/2})$ were anticipated to have very low photoassociation line intensities relative to the next high-lying vibrational level (three orders of magnitude lower than the $\nu = 2$ level) [23]. The attempt to measure (or, as we initially thought, only re-measure) the ‘missing’ vibrational levels was initially treated as a benchmark, but in the end, it allowed us to assess the quality of our experimental apparatus and its capabilities in detecting vibrational levels with extremely low intensities.

In this context, our experimental approach to measurements can be divided into two parts.

The first part can be considered as the enhancement of the photoassociation rate by using Feshbach resonances, namely Feshbach Optimized Photoassociation (FOPA) [26, 27, 28, 29, 30]. To facilitate this enhancement method, the initial states of the ultracold atomic samples must be well-known [31, 32, 33] and controlled via techniques that allow populating pre-determined spin states (spin-polarization). Our primary focus is spin-polarizing the ultracold cesium sample in the lowest possible energy state, $F = 3, M_F = 3$ (the procedure is detailed in Section 5.3). By populating the sample in a known spin state, the scattering length of colliding atoms can be tuned in a controlled way with Feshbach resonances. Since the photoassociation process relies on atoms colliding with each other, the ability to tune the collisional properties of atomic ensembles yields an enhancement in molecular formation rates [34] (refer to Section 5.4).

The second part of our approach aims to improve the accuracy and stability of detection. In the context of photoassociation, the ability to populate energy levels in the excited molecular potential is determined via observation of atom loss as a function of the frequency of the photoassociation laser, because whenever a molecule is formed, it results in a loss of two atoms. To increase the precision of measurements, we have employed a custom-made optical frequency comb [35](Section 4.6.2) that served as a frequency reference for the lasers used in the photoassociation experiments(Section 4.6). Regarding the detection stability of the measured loss of atoms, we aim to minimize readout fluctuations. In trap-loss spectroscopy, the quantitative assessment of the total number of formed molecules is determined either directly through photoionization [36, 37, 38] or indirectly by determining the remaining number of atoms [13, 39, 40]. In our setup, like in most modern experiments, we use the former approach. Since the initial atom number directly influences the final atom number, we have implemented a feature in our experimental control hardware that allows experimental sequences to be triggered by certain events, like, for example, reaching a certain fluorescence level during loading of a magneto-optical trap [41] (Section 4.5). This feature was employed in our apparatus to control the number of atoms prepared before the measurement phase by tracking the fluorescence signal of the atomic ensemble using a lock-in-based detection system (Section 4.4.3). By doing this, we ensured that the experimental sequence would progress to further cooling steps and to photoassociation measurements only upon reaching a certain number of atoms in the magneto-optical trap.

Thanks to these continuous developments, we have been able to validate the theoretical predictions made nearly 15 years earlier by N. Bouloufa et al. [23] by successfully observing the previously undetected lowest-lying vibrational levels in the external well of the $0_g^-(P_{3/2})$ state. Furthermore, our investigation revealed that previous experimental studies, which claimed to have been the first to observe these two lowest vibrational levels, probed neighboring vibrational energy levels belonging to other molecular states of Cs_2 . We have expanded our measurements to include the observation of these other molecular states, namely the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. In the past, these molecular states were only measured down to -120 cm^{-1} from the $(6S_{1/2} + 6P_{3/2})$ asymptote [42, 43, 25], while we were able to extend this range to -300 cm^{-1} , almost reaching an overlap with data collected in heat-pipe experiments.

The developed photoassociation setup has shown its effectiveness, providing us with the necessary tools and insights to conduct photoassociation measurements with high precision and an improved rate of molecule formation. In the near future, this particular setup will be used for forming bound KCs molecules. Additionally, photoassociation lasers and optical referencing tools will also be employed for studies of ultracold KAg and CsAg molecules within a research project that has been recently initiated.

As a guide for the reader, this thesis begins by presenting conceptual information on laser cooling of atoms in [Chapter 2](#), followed by a brief overview of ultracold molecules and the experimental methods for forming them in [Chapter 3](#). In this context, [Section 3.1.1](#) covers the direct laser cooling of molecules, while [Section 3.1.2](#) discusses photoassociation. Following this, [Section 3.2](#) presents information on molecular interaction potentials, particularly those related to the range of large internuclear distances. In [Chapter 4](#), one can find brief information on our K-Cs single-chamber and photoassociation apparatuses. [Chapter 5](#) presents the insights into the photoassociation measurements, their results, and their analysis. This chapter begins with our first attempts at probing excited molecular states in a magneto-optical trap in [Section 5.1](#), followed by measurements in an optical dipole trap in [Section 5.2](#). The latter sections give the details of the spin polarization of cesium atoms in [Section 5.3](#) and the utilization of Feshbach-optimized photoassociation for forming Cs_2 molecules in [Section 5.4](#), which has led to the observation of the two lowest vibrational energy levels of the external well of $0_g^-(P_{3/2})$ state in [Section 5.6](#) and extension of the photoassociation spectra of the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states in [Section 5.5](#). In the concluding [Chapter 6](#), a summary is provided, highlighting the contributions made and suggesting potential future projects that can be carried out with the constructed apparatus.

Chapter 2

Laser cooling and trapping of atoms

In ultracold physics, alkali atoms have played a pivotal role, giving rise to various laser cooling methodologies. In this thesis, our focus is on cesium, the heaviest stable alkali atom. We laser cool cesium atoms as part of our sample preparation phase for conducting ultracold photoassociation spectroscopy of cesium dimers. Cesium atoms have significance in establishing time standards through their ground state hyperfine splitting, making them pivotal for precise time measurements with atomic clocks [44]. Furthermore, cesium possesses a single valence electron that is loosely bound, resulting in a rich spectrum of electron-nucleus and atom-atom interactions. It is also known for its exceptional electro-positivity among all elements [45]; cesium boasts the largest polarizability and demonstrates the strongest van der Waals interaction strength [46].

In the context of producing ultracold cesium ensembles, the first successful implementation of a Magneto-Optical Trap (MOT) for cesium was accomplished by Monroe et al. in 1990 [47]. Since then, substantial progress has been made in decreasing the achieved temperatures and enhancing the densities of the cesium samples. Notably, spin-polarized cesium atoms were achieved a year later [33], and a few years following that, temperatures as low as $1.1 \mu\text{K}$ were attained through the application of gray molasses cooling (GMC) [48]. Subsequent years witnessed significant advancements in laser cooling techniques, which allowed the cesium atoms to be confined in optical dipole traps [49], the observation of Feshbach resonances [50], and the introduction of Raman sideband cooling [51]. Collectively, these innovations led to the achievement of the first Bose-Einstein Condensation (BEC) of cesium atoms in 2003 [52].

In this thesis, we employ several of these laser cooling techniques, specifically MOT, GMC, Zeeman pumping and the utilization of optical dipole traps, to cool and prepare ultracold cesium atoms for subsequent experiments, such as the formation of cesium dimers via photoassociation. The following sections introduce fundamental concepts in laser cooling and trapping, beginning with the MOT in [Section 2.1](#) and subsequently the widely employed sub-Doppler cooling technique, GMC, in [Section 2.2](#). The concept of optical dipole trapping is discussed later in the thesis, specifically in [Section 4.2.2](#), while the Zeeman pumping scheme is presented in [Section 5.3](#).

2.1 Magneto-optical trap

Light-matter interactions form the basis of laser cooling of atoms and molecules. When a photon interacts with an atom, not only does it transfer energy but also its momentum. Due to the conservation of momentum during atomic absorption, the atom gains or loses velocity depending on the direction of the photon relative to the motion of the atom. However, the absorbed energy is later emitted as a new photon during the spontaneous decay, which influences the momentum of the atom once more. In this scenario, the atom experiences a distinct ‘push’ in a well-defined direction (direction of the absorbed photon). Conversely, during spontaneous emission, the new photon is emitted in a random direction and as a result after many absorption and emission events the momentum transfer after emission average to zero. Thus, only the momentum lost due to the absorption effectively influences the net momentum change of the atom. In this context, if photons interact with atoms in a controllable manner, it becomes feasible to control the momentum of the atoms, thereby slowing them down to ultracold temperatures ($T_u < 1$ mK), which requires many interacting events between atoms and photons, as atoms at room temperature carry momentum orders of magnitude higher than the photons interacting with them.

These events are related to the scattering rate, referring to the rate at which an atom absorbs and re-emits incident photons. To achieve ultracold temperatures, typically 10^4 atom-photon interactions should happen. To achieve such a rate and thus a significant force on atoms, matching the frequency of photons with the optical transition of the atom is crucial. However, when employing this mechanism for cooling the atoms, the atom’s velocity must be considered due to the Doppler effect introducing frequency variations from the atom’s perspective. For example, when an atom moves parallel to the photon in the same direction, from the atom’s perspective, the frequency of the photons shifts to lower frequencies. In contrast, in the opposite direction, they exhibit higher frequencies. If perpendicular, it does not yield any difference. This phenomenon allows the selective targeting of specific groups of atoms based on their velocities, resulting in a mechanism that can exert velocity-dependent forces on atoms.

Two counter-propagating red-detuned beams (with frequency lower than the frequency of a transition) can be introduced to implement this mechanism. Velocity-dependent forces are applied to atoms via the Doppler shift, causing the counter-propagating beam to yield a higher absorption rate and exclusively lower absorption for the beam oriented in the same direction as the atom’s motion. Thus, the resulting total force on the atoms is favored to slow down the atoms. This velocity-dependent force system forms an atomic ensemble called optical molasses. While the atoms, in principle, can be slowed down significantly, the temperature achievable through Doppler cooling is constrained by the Doppler limit [45]:

$$k_B T_D = \frac{\hbar \Gamma}{2}, \quad (2.1)$$

where, k_B is the Boltzmann constant and Γ is the natural line-width of the cooling transition. While Doppler cooling methods can achieve temperatures in the 10’s of μK range, they fall short in providing high atomic densities since atoms are only cooled based on their velocities. For a cesium sample cooled on the D_2 line (upper level $P_{3/2}$), the Doppler limit is $125 \mu\text{K}$ [45]. To have colder and denser atomic ensembles, a trapping scheme is necessary, which draws the cold atoms to a specific location by introducing position-dependent force. This trapping condition

can be achieved by utilizing a magnetic field and cooling beams with appropriate polarizations. For example, a quadrupole field generated by coils with counter-flowing currents create magnetic field distribution with magnitude that increases radially from the center of the coils. The applied magnetic field induces Zeeman shifts of different spin states (M_F) of atoms [45]:

$$\Delta E = \mu_B g_F M_F |\vec{B}|, \quad (2.2)$$

where, μ_B is the Bohr magneton, g_F is the hyperfine Lande g-factor and $|\vec{B}|$ is the magnetic field amplitude. The selection rules for atomic transitions allow only σ^+ polarized photons to excite atoms from $|F, M_F\rangle \rightarrow |F' = F, F \pm 1, M_F + 1\rangle$, while σ^- is for $|F, M_F\rangle \rightarrow |F' = F, F \pm 1, M_F - 1\rangle$, and π is for $|F, M_F\rangle \rightarrow |F' = F, F \pm 1, M_F\rangle$. When two counter-propagating red-detuned beams with polarization orientations of σ^+ and σ^- are introduced, the exerted force on atoms becomes position-dependent due to the Zeeman shifts and the applied magnetic field variation along position (see Figure 2.1). This shift allows the σ^- beam to couple with the $M_F \rightarrow M_F - 1$ state when the atom is on the side of the incoming beam direction from the center of the magnetic field gradient. Thus, the atoms are pushed towards the magnetic field center. Conversely, the quantization axis, determined by the direction of the magnetic field, is reversed on the other side of the magnetic field center. Consequently, the σ^+ polarized beam couples with the $M_F \rightarrow M_F - 1$ state, pushing atoms toward the trap center again. It is worth noting that Doppler cooling is also utilized in this method, as the atoms still experience a force depending on their velocities. Together with the magnetic gradient and the laser beam polarization, the atoms are now subject to position-dependent force. So far, we have only covered two counter-propagating beams in one axis. Adding the same optical scheme to the other two Cartesian axes eventually traps the atoms in 3D, referred to as a 3D-MOT.

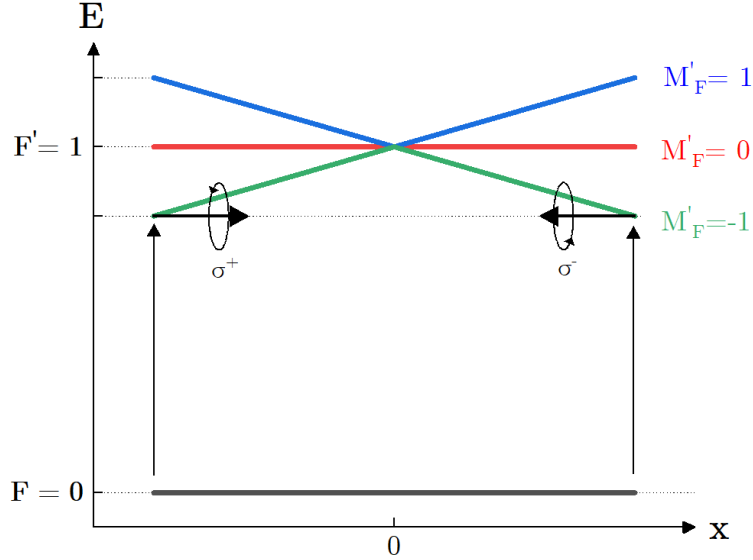


Figure 2.1: The Zeeman levels of an exemplary excited state $F' = 1$ is influenced by the spatial variation of the magnetic field along the optical axis x . Optical beams with σ^+ and σ^- polarizations are employed to excite atoms from the $F = 0, M_F = 0$ state to the $F' = 1, M'_F = -1$ state.

As mentioned earlier, to cool atoms from a hot background gas to ultracold temperatures, approximately 10^4 scattering events are required. As alkali atoms have a hyperfine structure in the ground state, excitation to an upper energy states can be followed by the decay to a state that is not a part of the cooling cycle, effectively reducing the population that can be efficiently cooled. To achieve the required level of photon scattering, the atom must undergo a closed cycling transition, ensuring that each photon absorption is followed by spontaneous decay back to the initial state. For states populated due to spontaneous decay, a ‘repumper’ laser becomes necessary to pump atoms back into the level that participates in the cooling process.

2.2 Gray molasses cooling

Gray molasses cooling (GMC) is a sub-Doppler cooling technique that involves the use of a blue-detuned cooling laser to drive atomic transitions of $F \rightarrow F' = F$ or $F \rightarrow F' = F - 1$ [48, 53]. This process induces the population of atoms into dark states, where their interaction with light significantly diminishes, resulting in reduced fluorescence (the term ‘gray’ signifies reduced fluorescence). This cooling method relies on a low photon scattering rate to reduce atom-atom interactions, thereby enabling the production of atomic samples at higher densities than conventional cooling methods such as MOT. However, typically, GMC cannot cool down the atoms from hot background gas; therefore, it is generally used following an operational MOT where the atoms are cooled down to temperatures where GMC can be utilized.

GMC relies on a Sisyphus effect between coupled states with spatially modulated light shifts and uncoupled states that experience no light shift. When a blue-detuned light with a spatially dependent polarization is illuminated on atoms, the dark states $|\psi_D\rangle$ do not experience an energy shift due to the present laser beam. However, the bright states $|\psi_B\rangle$ experience varying energy shifts induced by the polarization gradient of the laser beam.

As the atomic ensemble has a velocity distribution of atoms dependent on the temperature of the sample, when the atoms are in the bright state, their energies are being shifted increasingly. Thus, atoms are subject to losing kinetic energy as they gain potential energy. Once the potential energy of the atom increases, the chance of being excited to upper states with the blue-detuned light also increases. The atoms excited to the upper state will shortly decay either to the $|\psi_B\rangle$ or the $|\psi_D\rangle$ state in our two-level system. The decay to the $|\psi_B\rangle$ will lead to more kinetic energy losses, following an increase in the probability of being excited once again. However, if the atom decays to the $|\psi_D\rangle$ state, then atoms will have lower kinetic energies compared to the atoms in the $|\psi_B\rangle$ states. Figure 2.2 illustrates the process conceptually.

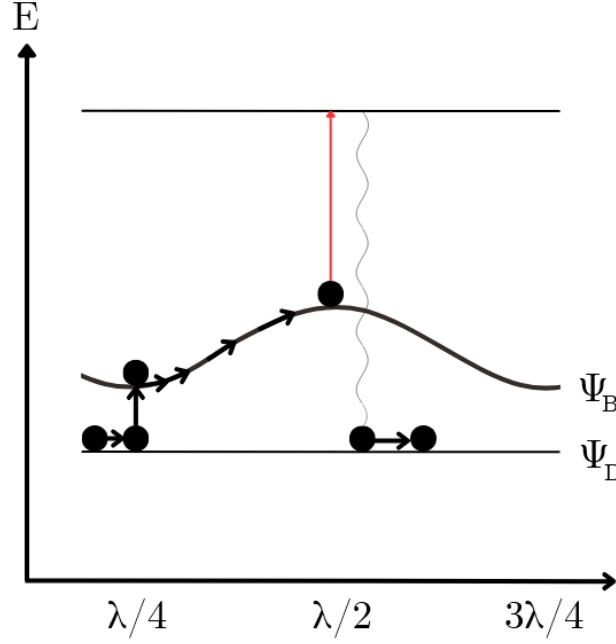


Figure 2.2: The figure illustrates the concept of polarization gradient cooling, where a blue-detuned light from the optical transition is present with spatially varying polarization. The polarization induces a splitting of the ground state into two states, $|\psi_B\rangle$ and $|\psi_D\rangle$. Atoms in the $|\psi_B\rangle$ state lose kinetic energy as they gain potential energy while climbing the potential caused by the polarization variation. Then, the blue-detuned light couples the atoms into the excited state. Spontaneous decay may put atoms into the $|\psi_D\rangle$ state, where the energy is lower. The transfer of atoms between the $|\psi_B\rangle$ state and the $|\psi_D\rangle$ state is velocity-selective.

An atom populating the dark state can transition to the bright state via velocity-selective coupling [54]. The probability of this process is proportional to the atom's velocity and inversely proportional to the light shift of the bright state. Any atom with a velocity other than zero will participate in the cooling cycle until reaching zero velocity. Atoms at rest will accumulate in the dark state. As the recoil from the last spontaneous-emission photon contributes to the cooling mechanism, the one-photon recoil energy is not the limiting factor [55]. In contrast, the resulting temperature is solely constrained by the coherent interaction time [56]. However, the imperfections introduced in a real-life implementation of this cooling method define a practical temperature limit, which often can be a few factors above the recoil temperature ($T_{\text{recoil}} \approx 198$ nK for cesium). Similarly to the principles of the MOT, spontaneous decay to levels other than the initial states is possible. Thus, the atoms in other states are pumped back into the optical cycle via a repumping transition.

Chapter 3

Ultracold molecules

In the preceding chapter, we discussed the techniques used to cool cesium atoms from a hot background gas to temperatures as low as a few tens of microkelvin. These methods can be directly extended to molecules, facilitating the direct determination of their properties without the necessity of forming ultracold molecules via laser-cooled atoms. This chapter aims to provide an overview of the methods employed in realizing ultracold molecules, presenting readers with a comparative perspective. Additionally, brief information regarding molecular states, such as interaction potentials that depict interatomic forces within a molecule, information on molecular states, and molecular selection rules are presented.

It should be noted that the information provided has been guided by the Refs. [14, 57, 58, 59], which have been significant sources of knowledge for this thesis work.

3.1 Formation of ultracold molecules

3.1.1 Direct laser cooling

In recent years, a notable effort has been made to apply laser cooling methods, developed initially for atoms, to molecules. The utilization of laser cooling methods on atoms has led to extremely low temperatures [60, 61, 62]. The effectiveness of laser cooling primarily arises from the continuous scattering of photons by molecules (or atoms) when exposed to a laser. Achieving ultracold temperatures, as discussed earlier, demands approximately 10^4 photon scatters, requiring a closed cycling transition for consistent absorption and spontaneous decay [18, 63].

Unfortunately, in most species, be it atoms or molecules, close transitions are usually absent, with some notable exceptions like metastable ^4He [64]. Spontaneous decays to other states inevitably occur, typically before reaching ultracold temperatures. For these other states that are populated due to spontaneous decays, a ‘repumper’ laser becomes necessary to pump these populated states back into the main cycle. This enables the continuation of photon scattering, eventually cooling the systems to the ultracold regime. While atomic systems commonly feature cycling transitions

that require one or two repumper lasers, finding such transitions in molecules is challenging due to their vibrational and rotational degrees of freedom. Managing vibrational states poses a challenge since there is no strict selection rule governing the ratios for the decay of an excited electronic state into different vibrational levels [15, 18]. Such decay to different states requires more lasers to pump the population back into the main cycle, presenting an experimental challenge. Another issue arises from the susceptibility of molecules in rovibrationally excited states to collisional relaxation. When these molecules undergo collisions, there is a high likelihood of transitioning to lower rovibrational levels, accompanied by the release of energy that elevates the kinetic energy of the molecules. This phenomenon commonly leads to undesired outcomes such as heating and trap loss, posing a significant constraint on preparing dense and ultracold molecular samples [65].

Despite these experimental challenges, in many aspects, the development of laser cooling for molecules has followed a path similar to that pioneered with atoms several decades ago [57]. Initially, a beam of molecules underwent laser cooling through a transverse cooling method for molecules, such as SrF [18] and YO [63], followed by further cooling through radiation pressure [66]. Subsequently, magneto-optical trapping was utilized for a few molecular species, including CaF [16, 67], SrF [68], and YO [69]. Sub-Doppler cooling methods were employed to bring these trapped molecules into the ultracold regime [16, 67, 70]. For some species, temperatures as low as a few microkelvins have been reached. For example, 100 μ K was achieved with YbF [71], 5 μ K with CaF [72, 73], and 4 μ K with YO molecules [74].

Following a similar trajectory to atoms, these molecules were confined in magnetic traps [75], optical dipole traps [76], and optical tweezers [77]. Their internal states were controlled using microwave fields for CaF [75, 78] and RbCs [78] molecules. Continuing, the collisional properties of molecules were investigated, both among laser-cooled molecules and between laser-cooled atoms and molecules [79].

These direct laser cooling methods and control schemes have been described for diatomic molecules so far. Recently, there has been progress in extending these methods to polyatomic molecules, such as CaOH in a one-dimensional MOT with temperatures in the few millikelvins (2-8 mK) range [80], and Sisyphus laser cooling of CaOCH₃ cooled down the molecules to sub-millikelvin temperatures [81].

It is worth mentioning that direct laser cooling techniques are predominantly utilized on alkaline-earth fluoride compounds [82], as well as a limited number of molecules that meet specific prerequisites. As mentioned earlier, this criteria includes having a specific energy level structure and Franck–Condon factors between vibrational energy levels of the electronic ground state and vibrational levels of the first electronic excited state, allowing mainly for the so-called ‘diagonal transitions’ and enabling the realization of a closed optical cycling transition [83].

Ultracold molecules can also be cooled further down once formed from ultracold atoms. Several molecules assembled from ultracold atoms have been successfully created in the rovibrational singlet ground state. These include LiK [84], NaRb [85], NaCs [86], KRb [87], Na⁴⁰K [88, 89, 90], Na³⁹K [91], and RbCs [92, 93]. Additionally, in the ground triplet state, molecules such as Li₂ [30], and LiNa [94] have also been achieved.

In a subject closely related to this thesis, it is crucial to note that there has been no reported progress on the direct laser cooling of Cs₂, K₂, and KCs molecules. However, advancements have been made in cooling Cs₂ and K₂ molecules formed from already cold atomic ensembles.

A notable example involves shifting the population to lower vibrational levels using a specially tuned light source for Cs_2 [95] and K_2 [96]. Moreover, they were used in employing techniques involving Feshbach association on Cs_2 [97, 98], on K_2 [99, 100] and on KCs [101] molecules. Additional cooling methods were introduced on Cs_2 molecules [102] and for KCs molecules [103] via STIRAP (Stimulated Raman Adiabatic Passage). STIRAP is a technique used in quantum physics to efficiently transfer the population between two molecular quantum states without loss. Here, the system is manipulated with a pair of laser pulses whose frequencies are carefully chosen to drive transitions between different molecular states. By adjusting the timing and intensity of these laser pulses, the population can be smoothly transferred from an initial to a final state while minimizing the population in intermediate states.

As progress on the direct laser cooling of molecules expands to other species of molecules, the importance of alternative techniques, such as photoassociation and the creation of Feshbach molecules, will remain. These techniques are recognized for their adaptability and potential to complement direct cooling methods. However, it is essential to acknowledge that assembling molecules from ultracold atoms is challenging, potentially as difficult, or even more so than direct cooling. While these techniques enable the creation of high phase-space density molecular gases, achieving this goal involves overcoming significant technical challenges. For instance, establishing a Bose-Einstein condensate (BEC) and a degenerate Fermi gas in the same setup, forming Feshbach molecules, or implementing techniques like STIRAP with their complex laser systems are formidable tasks.

3.1.2 Photoassociation of ultracold atoms

One of the methods for forming ultracold molecules from ultracold atoms is ultracold photoassociation. Photoassociation occurs through the absorption of resonant light by two colliding atoms, resulting in their photoassociation and the formation of a bound, electronically excited molecule:



This process occurs only when the total kinetic energy of the atoms, combined with the energy of the absorbed photon, matches the energy of a bound molecular state. Once the electronically excited molecule is formed, it quickly decays by emitting a photon. During this decay process, the energy of the emitted photon determines the final product, which can either be two free atoms or a bound molecule in the ground electronic state. Therefore, the excited state molecules are relatively short-lived. The decay process can be shown as follows (Equation 3.2 in the case where two free atoms are formed or Equation 3.3 if the final product is a ground state molecule):



If the decaying photon's energy $h\nu_d$ is lower than the initially absorbed photon's energy $h\nu$, the resulting two atoms in the final product will have higher kinetic energy than the initial kinetic

energies. However, it can also be lower or the same depending on the energy of the decaying photon. In the ultracold samples, the variation in the initial kinetic energies of colliding atoms is minimal. Consequently, the range of photon frequencies required to drive the resonant photoassociation process is also limited. This frequency range can be comparable to or even smaller than the natural width of the electronically excited molecular state [14]. It is also possible to observe photoassociation at elevated temperatures [104]; however, achieving high spectral resolution under such conditions is not possible when compared to samples at ultracold temperatures. Additionally, the methods developed for ultracold atoms allow the selection of internal states with methods such as optical pumping [32]. Utilizing precise control over the initial conditions of prepared ultracold atomic ensembles is advantageous for favoring the photoassociation of ultracold atoms, as it leads to the formation of molecules through the transition of their initial states to the resulting molecular states.

The measurements that utilize the processes mentioned above, known as ultracold photoassociation spectroscopy, allow the study of molecular states by forming molecules at extremely low temperatures. This provides insights into their properties and behaviors, particularly at large internuclear distances. At short internuclear distances, overlapping electron clouds lead to effects requiring complex descriptions of molecular wave functions. Conversely, at long-range internuclear distances, undistorted electron clouds allow for a simpler tracing of molecular properties, such as spin-orbit and hyperfine structure. Ultracold photoassociation spectroscopy unveils these molecular properties such as ground state molecular dissociation energies [9, 10] and, in the case of alkali dimers, it can also be used for calculating the radiative lifetimes of the atoms [23, 105, 106].

The main focus of this thesis revolves around a specific state, the external well of the $0_g^-(P_{3/2})$, which is a long-range state. This allows the analysis of experimental data for the external well of this state within theoretical models developed solely for long-range interactions [19]. This state is often called a purely long-range state, where all rovibrational energy levels of a molecule are situated at long-range internuclear distances. In this thesis, we do not use such terminology since this state has a double-well structure with vibrational levels in its inner well out of the long-range interval, which can be probed via tunneling from the external well to the inner well [107, 108]. Also, exciting atoms in the triplet ground state can populate the inner vibrational levels [102]. To our knowledge, there is no available spectroscopic information for the inner well apart from these narrow-range measurements. However, their presence disallows this state to be called as ‘purely’ long-range.

3.2 Interaction potentials

Photoassociation not only enables the creation of new molecular entities but also provides a unique window into the underlying interaction potentials governing this formation of molecules. The interaction potentials, representing the interatomic forces, play a crucial role in determining the outcome of the photoassociation process.

In this section, brief information on molecular states is given, together with a short description of Hund’s cases (a) and (c). In conventional molecular spectroscopy, where the focus is on alkali dimers within an internuclear distance approximately shorter than 10 Å, the emphasis is on electronic states characterized by Hund’s cases (a) and (b). Beyond this range, specifically at distances around more than 20 Å, ultracold photoassociation spectroscopy primarily examines

electronic states governed by Hund's case (c) [59].

In this thesis, the vibrational levels of the states $0_g^-(P_{3/2})$, $1_g(P_{3/2})$ and $0_u^+(P_{3/2})$ are probed, and these states are described by Hund's case (c) (see Figure 3.1). While it is described by Hund's case (c), they are a result of mixing between two Hund's case (a) electronic states; $^3\Sigma_g^+(6s+6p)$ and $^3\Pi_g(6s+6p)$.

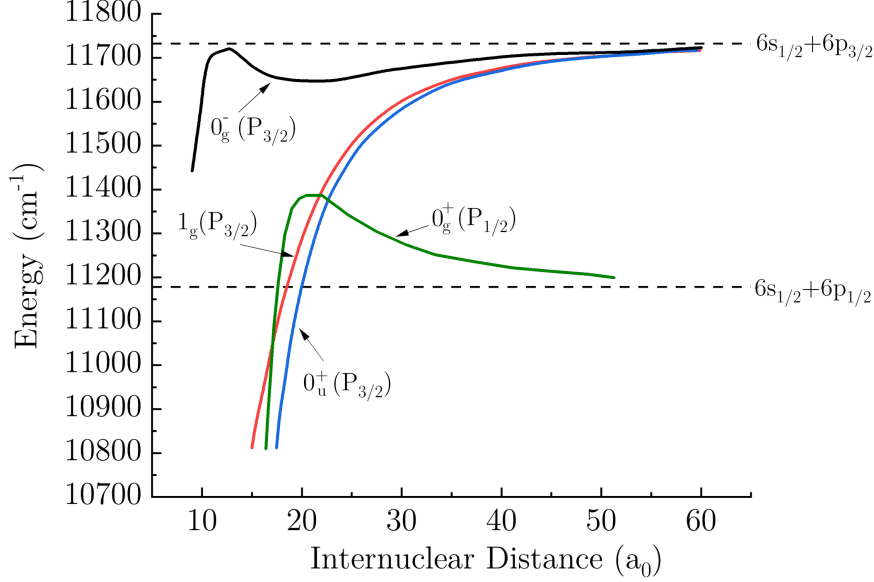


Figure 3.1: The Hund's case (c) potentials, which are analyzed and mentioned in this thesis. $0_g^-(P_{3/2})$ (black), $1_g(P_{3/2})$ (red), $0_g^+(P_{1/2})$ (green) and $0_u^+(P_{3/2})$ (blue) of Cs_2 molecule. The potential energy curves were obtained through digitization from Refs. [23, 42, 109].

The external well of the $0_g^-(P_{3/2})$ as shown in Figure 3.1 is characterized by a potential where the Coulomb force dominates interactions. As the internuclear distance decreases, the interaction between atoms is primarily influenced by the overlap of electron orbitals. These long-range interaction potentials can be described with the formulation presented in Equation 3.4. The condition of a long-range potential is defined by the LeRoy radius (R_c). If the outer turning point of vibrational levels $R_{\nu+}$ exceeds R_c , the interaction between the atoms is assumed to take a long-range form [110, 111]. This statement arises due to the fact that when the molecule vibrates within a potential well, spanning from an inner turning point to an outer turning point, it predominantly resides in the long-range region. In the long-range region, the interaction between the two atoms can be expressed as a series of terms depending on powers of $1/R$:

$$V(R) = D - \sum_n C_n R^{-n}. \quad (3.4)$$

In Equation 3.4, R is denoted as the internuclear distance, and D represents the dissociation limit, signifying the energy threshold beyond which the molecule can no longer exist, meaning the pair of atoms can no longer be bound together. In Table 3.1, one can find the dispersion coefficients C_n and their cause of contribution to the interaction potential.

Table 3.1: The dispersion coefficients contributing to the interaction potential given by Equation 3.4 with the cause of interaction between atoms given.

Coefficient	Interaction
C_1	Ion-Ion
C_2	Ion-Dipole
C_4	Charge Induced Dipole
C_3	Dipole-Dipole (Resonance)
C_5	Quadrupole-Quadrupole
C_6	Induced Dipole
C_8	Dipole-Quadrupole

These coefficients are obtained by fitting experimentally obtained data into theoretically derived interaction potentials. In this thesis, various theoretical approaches are analyzed to determine these interaction potentials. The choice of theoretical method for fitting depends on the specific challenges posed by the behavior of the interaction potentials. Different methods are utilized based on the required accuracy for a particular fit, and these models are developed to address challenges arising from the specific characteristics of the interaction potentials. As an example of the usage of different models, when the internuclear separation is close to the LeRoy radius, the overlap between the two atomic charge distributions starts to influence the potential. Therefore, the interaction potential should also include the exchange effects between the two atoms. This exchange effect is derived from the asymptotic exchange interaction proposed in Ref. [112]. In the latest revision of the $0_g^-(P_{3/2})$ state [23], a preference is given to the analytical asymptotic approach over the Rydberg–Klein–Rees (RKR) method [113, 114, 115]. This choice is attributed because the inner barrier of the external well, which is a result of mixing by spin-orbit interaction of two asymptotic potentials converging toward the $6S_{1/2} + 6P_{1/2}$ and $6S_{1/2} + 6P_{3/2}$ limits, is not as steep as expected in the RKR model. Consequently, the previous theoretical approaches yielded inconsistent results when applied to the analysis of the experimentally obtained data.

Additional comments on the interaction potentials and theoretical models for molecular states are discussed later in this thesis, specifically for the states $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$, which are provided in Section 5.5, and for $0_g^-(P_{3/2})$ in Section 5.6.

3.2.1 Labelling of molecular states

In the context of molecular states, they are labeled by the molecular term symbol, exemplified in the case of diatomic molecules by:

$$^{2S+1}\Lambda_{g,u}^{\pm}. \quad (3.5)$$

Here, S represents the total electronic spin quantum number, and Λ is the quantum number describing the projection of the total electronic angular momentum onto the internuclear axis. The symbols $\pm$ denote the symmetry with respect to the plane containing nuclei, while the labels u/g indicate the behavior of the wave function upon the exchange of nuclei. Molecular states are distinguished by a combination of a letter and a molecular term symbol, as given above, where the

letter signifies the relative energy order of electronic states, and the term symbol communicates their symmetry properties. The ground state, the energetically lowest, is universally denoted as X. Sequentially higher-energy states of the same spin multiplicity as the ground state are labeled A, B, and so forth. States of other multiplicity (or multiplicities) are designated with lowercase Roman letters, commencing with "a" for the lowest energetic state, "b" for the subsequent higher state, and so forth.

Refining the classification of an electronic state becomes necessary when the impact of the spin-orbit interaction cannot be neglected. In such instances, it proves beneficial to discern between two distinct scenarios: weak and strong spin-orbit interactions. From these approaches, the Hamiltonian of the system can be defined, and the rovibrational energy levels can then be estimated theoretically. Typically, experimental data is fitted to theoretical models using fitting tools. By employing these tools, which yield the "best" results, the parameters that define the model are determined.

3.2.2 Hund's case (a)

In Hund's coupling case (a), the electronic orbital angular momentum is coupled to the internuclear axis by electrostatic forces, and the electronic spin angular momentum is coupled to the electronic angular momentum by spin-orbit coupling. The coupling of the electronic orbital angular momentum to the internuclear axis is stronger than the spin-orbit coupling; hence, the effect of spin-orbit interactions is weak.

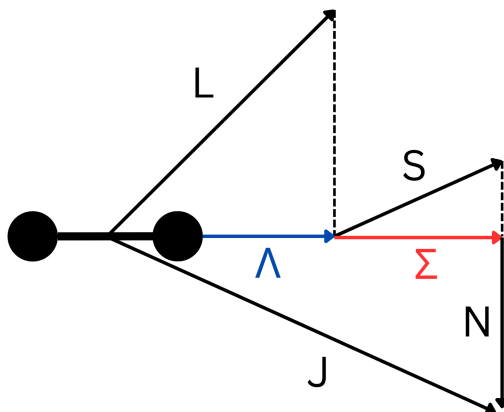


Figure 3.2: Illustration showing the Hund's case (a).

When one considers the orbital angular momentum of electrons L , the projection of L onto the internuclear axis will define the quantum number Λ . Thus, Λ can take values of 0, 1, 2, $\dots$, L . These quantum numbers are often labeled with Greek letters of Σ , Π , Δ , Φ , and so on.

A similar approach to the spin-orbit coupling can be done where the spins of the electrons couple to a total electron spin defined by S . The projection of S on the internuclear axis is denoted as Σ , where in this case, Σ can have values ranging from $-S$ to S , incrementing by one unit, resulting in a total of $2S + 1$ different values being present. When both projections are added up $\Omega = |\Lambda + \Sigma|$, it will yield the total angular momentum of the electrons.

Moreover, the molecule of interest can rotate around an axis perpendicular to the internuclear axis, as illustrated in [Figure 3.2](#). The angular momentum of this rotational movement is denoted as N . Then, the total angular momentum J of the molecule becomes the sum of the angular momentum of the electrons Ω and N . Considering that the rotation axis is to be perpendicular to the internuclear axis, the total angular momentum J can only be equal to or higher than the Ω . The total rotational energy of the molecule is often well resolved in the photoassociation spectroscopy and the corresponding energy of the rotational energy levels is

$$E_{rot} = B_{rot}[J(J+1) - \Omega^2]. \quad (3.6)$$

B_{rot} is the rotational constant corresponding to a given vibrational energy level. In the case of $\Lambda = 0$, the rotational energy is slightly modified to be

$$E_{rot} = B_{rot}K(K+1), \quad (3.7)$$

where, it represents Hund's case (b). The electron orbital angular momentum, represented by Λ , couples with the molecular rotation N to yield the total momentum K . Simultaneously, the electronic spin couples with K , resulting in the formation of the total angular momentum J .

The necessity for this slight change arises due to the fact that the Σ states do not have contributions from spin-orbit coupling. In [Equation 3.7](#), K is given as $K = |\Lambda + N|$. In this case, the total angular momentum J can have the values from $J = (N + S), (N + S - 1), \dots, |N - S|$. In the context of this thesis, Cs_2 as an alkali dimer can only have $S = 0, 1$, hence in the case of $\Omega = 0$, it can only have the states $^1\Sigma$ or $^3\Sigma$ [13]. For the $^1\Sigma$ state, J is equal to N , and the rotational energy is solely determined by the rotation of the molecule around its axis. In the case of the $^3\Sigma$ state, J can take values of N or $N \pm 1$, and a rotational energy level with a certain energy corresponding to one N can be composed of several different values of the total angular momentum J .

3.2.3 Hund's case (c)

In Hund's case (c), the spin-orbit coupling becomes dominant over the coupling of the angular momentum to the internuclear axis. Thus, the Λ projection of orbital angular momentum L on the internuclear axis and the Σ projection of the total electron spin on the internuclear axis become undefined.

In such a case, the angular momentum of the electrons is denoted as J' . J' is subsequently coupled to the molecular axis, yielding the well-defined component Ω . The electronic angular momentum, Ω , and the angular momentum of nuclear rotation, N , then combine to form the total angular momentum of the system, J (see [Figure 3.3](#)).

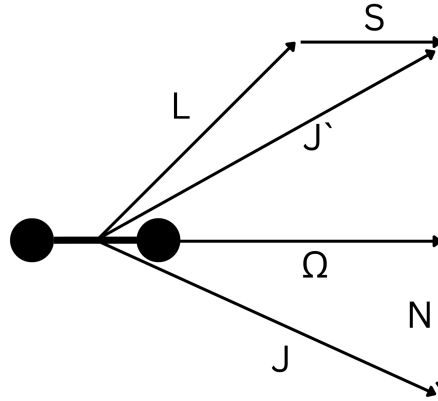


Figure 3.3: Illustration showing the Hund's case (c).

In the long-range interactions, the states are labeled as:

$$\Omega_{u,g}^{\pm}, \quad (3.8)$$

where, the superscript denotes the symmetry with respect to the molecular plane and the subscript denotes the behavior of the wave function at the exchange of the nuclei, these states can be, for instance, given as the states $0_g^-, 0_u^+$ and 1_g . The concept of u/g symmetry is exclusively defined for homonuclear molecules, where identical nuclei contribute to the symmetry operations.

3.2.4 The selection rules

The selection rules governing the states of interest provide insights into the possible decay pathways for molecules formed through the process of photoassociation [14]. These selection rules prohibit changes in the total angular momentum by more than one unit, restricting transitions from $J = 0$ to $J' = 0$. From further selection rules given in Table 3.2, the transitions from a singlet state to a triplet state are forbidden [116].

Table 3.2: Selection rules for molecular transitions given for homonuclear molecules [116].

Property	Allowed Changes
Total Angular Momentum	if $\Omega = 0 \rightarrow \Omega = 0$ then $\Delta J \neq 0$
Total Electron Angular Momentum	$\Delta\Omega = 0, \pm 1$
Projection of Electron Spin	$\Delta\Sigma = 0$
Orbital Angular Momentum	$\Delta\Lambda = 0, \pm 1$
Electron Spin S	$\Delta S = 0$
Parity:	$g \leftrightarrow u, g \not\leftrightarrow g, u \not\leftrightarrow u$
Symmetry:	$\pm \leftrightarrow \pm, + \not\leftrightarrow -, - \not\leftrightarrow +$

The transitions between rovibrational levels of the $0_g^-(P_{3/2})$ state and $a^3\Sigma$ state rovibrational

levels follow the selection rules according to which J changes by ± 1 or stays the same. In the $a^3\Sigma$ state, the lowest rotational level has the projection of the electron orbital angular momentum coupling with the molecular rotation N equal 0 ($K = 0$). Thus, the rotational levels of the $a^3\Sigma$ can have the values of $J = K, K \pm 1$. The selection rules from J to J' were stated above. For instance, a rotational level $(\nu, J) = (\nu, 0)$ in the state $0_g^-(P_{3/2})$, can decay to $K = 0, 1, 2$ in the $a^3\Sigma$ potential. This specific potential was used to create the first triplet ground-state molecules of Cs_2 and while $1_u(P_{3/2})$ state in the formation of molecules in the last bound levels of the singlet state [117]. The transitions $0_g^- \rightarrow a^3\Sigma_u^+$ are observable, but $0_g^- \rightarrow X^1\Sigma_g^+$ cannot be observed due to the fact that Hund's case (a) $^1\Sigma_g^+$ state corresponds to Hund's case (c) 0_g^+ whereas Hund's case (a) $^3\Sigma_u^+$ state corresponds to Hund's case (c) 0_u^- and 1_u . In this case, transitions $0_g^- \rightarrow 0_g^+$ are doubly forbidden (as g to g and $+$ to $-$) whereas $0_g^- \rightarrow 0_u^-$ and $0_g^- \rightarrow 1_u$ are allowed. Furthermore, transitions from $0_u^+ \rightarrow X^1\Sigma_g^+$ are also allowed and have been used in a STIRAP scheme for creating ground state molecules [118].

Chapter 4

Experimental setup

Now that we have provided conceptual information on methods for laser cooling of atoms, molecules, and molecule formation at ultracold temperatures, we can proceed with detailing our experimental apparatus.

Experimental setups used for the investigation of ultracold atoms and molecules consist of highly complex optical and electronic apparatuses. These devices generally require a fairly stabilized and consistent operation. In this chapter, one can find a brief description of these devices, which are built to prepare ultracold samples of cesium and potassium mixtures to form ultracold ground-state KCs molecules in our setup. However, this thesis primarily focuses on cesium atoms and the formation of excited Cs_2 molecules via photoassociation. Therefore, only aspects of the setup relevant to the laser cooling and manipulation of cesium atoms will be discussed. Details of the apparatus relevant to potassium will be covered in the future doctoral thesis of Mateusz Bocheński [119].

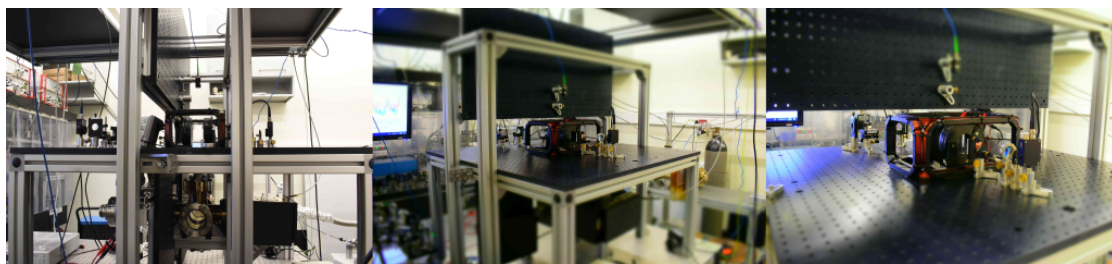


Figure 4.1: Single Chamber K-Cs setup in its early development stages (as of 2019). The image was captured after the vacuum chamber assembly and the installation of magnetic coils.

The potassium laser system is constructed in such a way that all three stable isotopes of potassium (^{39}K , ^{40}K , and ^{41}K) can be studied, including their mixtures. As a result, our experimental setup allows us to investigate ^{39}K -Cs, ^{40}K -Cs, and ^{41}K -Cs ultracold molecules. Both potassium and cesium atoms are released from commercially available dispensers made by SAES Getters (AMD), located 10 cm from the trapping region. These dispensers are manufactured with a material that

has sufficiently high resistance, causing it to heat up to several hundred degrees Celsius upon running a current of 3.5 A. Consequently, the hot cesium and potassium atoms are released into the experimental chamber, which consists of a glass cell. The glass cell ensures easier optical access, which is crucial for operating two species. The released hot atoms are initially laser-cooled down to 100 μ K inside this glass cell, and further cooling methods and experiments are then conducted within the same glass cell.

Our light preparation setup for cooling cesium atoms is designed to generate the essential light beams for successive stages in the cooling and confinement process. This includes the operation of the Magneto-Optical Trap (MOT), Gray-Molasses-Cooling (GMC), dipole trap confinement, spin-polarization, and imaging of the prepared samples. A detailed description of the cesium light preparation setup can be found in [Section 4.2.1](#), and for the optical dipole trapping setup, refer to [Section 4.2.2](#). The imaging of the cesium samples is discussed in [Section 4.4](#), and the spin-polarization is detailed later in [Section 5.3](#).

The single-chamber experimental setup, as introduced in the earlier sections, has yielded results with high-precision photoassociation measurements of cesium atoms. Notably, it distinguishes itself as the first apparatus capable of detecting numerous photoassociation lines that had not been observed previously.

As mentioned in [Chapter 1](#), although our photoassociation measurements have produced unique results, its distinction from other photoassociation setups does not arise from any novel methods. It is crucial to acknowledge that the capability to conduct cesium photoassociation studies is not exclusive to our group. Many research groups working with cesium possess or have had experimental setups capable of conducting such measurements. Other research groups may not have pursued these studies with the same level of precision and attention to detail while utilizing their experimental apparatuses. In the following sections, we introduce the experimental apparatus for cooling cesium atoms to ultracold temperatures and conducting high-precision photoassociation spectroscopy of cesium atoms.

This chapter will start with brief information about our vacuum chamber in [Section 4.1](#) following the optical design of the cesium cooling lasers ([Section 4.2.1](#)) in which details of the laser beams tailored for specific laser cooling methods are given. The following section ([Section 4.2.2](#)) covers the laser system for confining the atoms via optical dipole force.

Apart from our optical setups, we utilize magnetic field sources, detailed in [Section 4.3](#). This section covers the characterization of the electromagnetic coils we have designed and constructed for the KCs single chamber setup and our future research projects.

[Section 4.4](#) details the imaging setup where, our imaging objectives are introduced with their design and characterization. Imaging methods such as absorption imaging and fluorescence imaging are also discussed in this section. Additionally, we utilize a lock-in-based detection scheme for readouts of fluorescence signals emitted by our ultracold atoms. [Section 4.4.3](#) gives insights into the assembled electronic scheme for conducting lock-in-based readouts for both species.

Our experimental control hardware is introduced in [Section 4.5](#) and in the final section of this chapter ([Section 4.6](#)), assembly of two titanium-sapphire lasers is given together with their distribution and optical referencing to an optical frequency comb.

4.1 Vacuum systems

We begin the experimental setup chapter with our fundamental component, the vacuum chamber. The construction of the vacuum system was primarily led by Paweł Arciszewski, with comprehensive details provided in his master's thesis [120]. Nevertheless, my involvement in the construction process was comprehensive, spanning from the cleaning of parts to the assembly and bakeout stages.

Our ultra-high vacuum chamber (illustrated in Figure 4.2) comprises a rectangular UV fused glass cell connected to a 6-way CF4.5 stainless steel cross via a standard glass-to-metal transition. The glass cell is manufactured by 'Precision Glassblowing (Denver, US)' and does not have anti-reflection coatings. The six-way cross below the glass cell serves as a foundation for interconnecting various components of the vacuum chamber, three of which are window ports with anti-reflection coatings. The lower port is designated for vertical absorption imaging, while the other two facilitate the analysis of the ultracold sample after allowing it to fall to the six-way cross under gravitational force. However, only the port intended for vertical imaging has been utilized in the experiments conducted thus far.

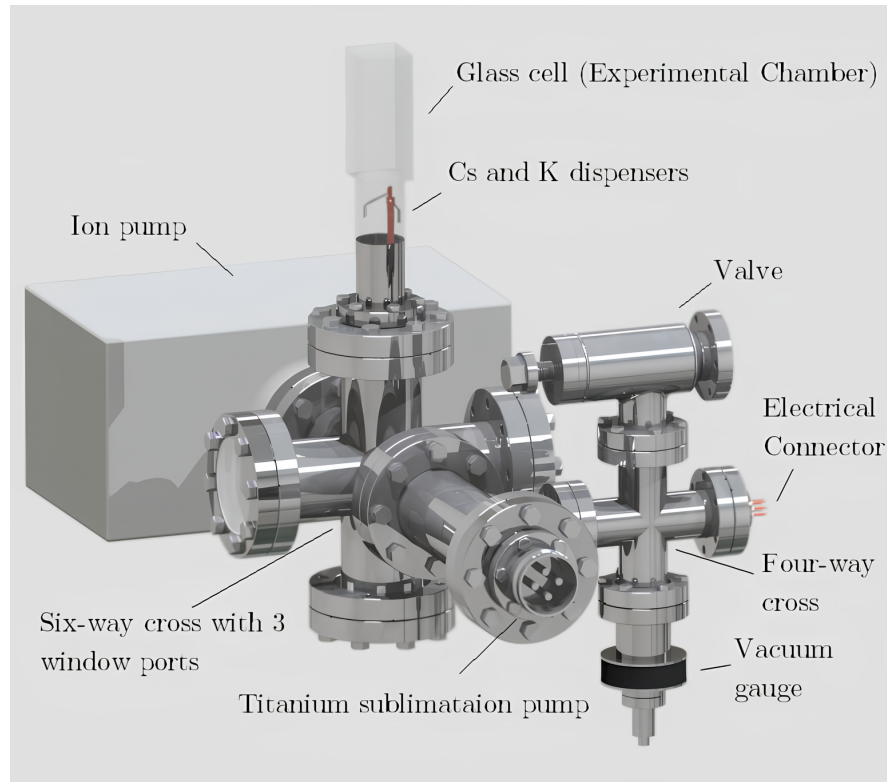


Figure 4.2: The design of the vacuum chamber is presented through 3D modeling software, highlighting its elements and positions. Figure taken from Ref. [120].

Two pumping units are employed to maintain ultra-high vacuum conditions within the chamber.

The primary pump is an ion pump (*Agilent Technologies VacIon Plus 75*) with a pumping speed of up to 75 l/s. Additionally, a titanium sublimation filament (*Agilent Technologies Titanium Sublimation Cartridge*) is positioned on the opposite side of the cross from the ion pump. Adjacent to the sublimation pump, a smaller four-way cross features slots for essential elements, including an inverted magnetron vacuum gauge (*Agilent Technologies IMG-300*), a main valve, and an electrical feedthrough. The electrical connector plays a crucial role in facilitating the flow of currents through our cesium and potassium dispensers.

These dispensers, four in total, are integrated into our setup, with two dedicated to cesium and two dedicated to potassium. They are mounted on a copper pole to position them in proximity to the trapping region (see [Figure 4.2](#)). In typical operation, a current of 3.6 A flows through the cesium dispensers and 3.8 A through the potassium dispensers. To minimize any potential impact on the cooling process of atoms, we have implemented a MOSFET-based electronic switch, allowing us to control the current and turn it on and off during critical stages of the experiment. To determine the magnetic field created by each active dispenser, we applied the Biot-Savart law. The dispenser was modeled as a finite-length wire inclined at a 45° angle with respect to the vertical axis and positioned approximately 10 cm away from the trapping region. In typical operation, this configuration results in a magnetic field measuring approximately 40 mG at the location of the atoms for each active dispenser with 3.6 A of current flowing through the dispenser.

The vacuum system is supported by an aluminum frame, as illustrated in [Figure 4.1](#). This frame is intentionally designed to accommodate horizontal and vertical optical breadboards, providing convenient space for placing optical components around the glass chamber. Constructed using $45 \text{ mm} \times 45 \text{ mm}$ T-slot profiles, the aluminum frame ensures flexibility in configuring the setup. The vertical profiles are filled with lead beads to enhance stability and dampen vibrations. Importantly, all components within this framework are non-magnetic to minimize the possibility of magnetizing the frame during the quick turn-on and off of the magnetic coils. Additionally, we reduce the impact of varying stray magnetic fields by refraining from positioning components made of magnetic materials.

4.2 Laser systems

This section discusses our laser systems essential for the cooling, manipulation, and confinement of cesium atoms within optical dipole traps and lattices. Paweł Arciszewski played a pivotal role in constructing the experimental setup, particularly in cesium laser frequency stabilization and designing tailored beams for various experimental techniques. Expanding upon this framework, I led the integration of an additional component, focusing on spin polarization for our cesium ensemble. Additionally, I took on the responsibility of maintaining the setup and optimizing all associated processes.

4.2.1 Cesium cooling lasers

The cesium laser setup is designed to ensure stable light generation and precise control over its properties. The laser light preparation setup is divided into two distinct parts. The first part

is dedicated to the light generation and frequency stabilization of the lasers positioned on an isolated optical table. This separation ensures the stability of the lasers without interfering with the ongoing activities in the laboratory so that the lasers remain undisturbed. The second part is focused on tailoring the light frequencies to prepare beams for additional cooling and control sequences of atoms. This second setup is positioned closer to the KCs single chamber apparatus for convenient distribution and alignment of the laser beams toward the experimental chamber.

The initial light generation is achieved using two external cavity diode lasers (ECDLs) by ‘*Toptica Photonics*’, as depicted in Figure 4.3. In this thesis, we use the terms ‘*cooling laser*’ and ‘*repumper laser*’ for these two lasers that are used to denote their initial purpose in operating the MOT.

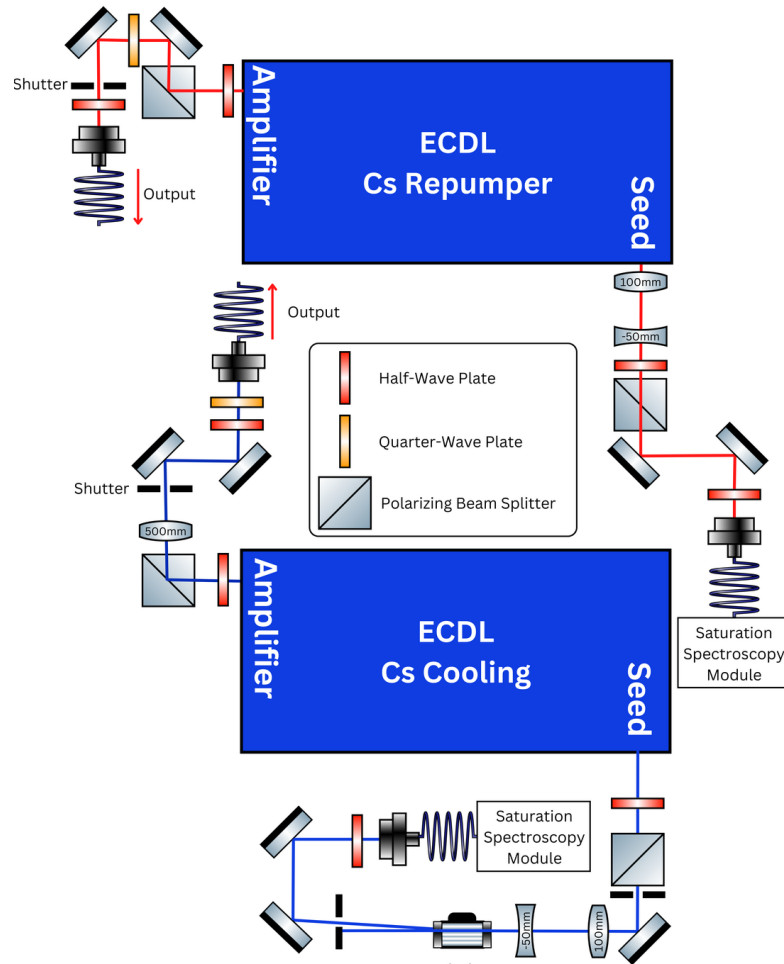


Figure 4.3: Optical layout of the cesium repumper laser and cesium cooling laser generation and distribution setups. A small portion of the seed light is allocated for stabilization and, therefore, directed to a saturation spectroscopy module. The remaining seed light is routed to the amplifier within the same housing as the laser modules. The amplified laser light is subsequently directed to the optical table in the vicinity of the KCs single-chamber apparatus for further refinement. *The optical components depicted in this figure are licensed under [121].*

However, during various cooling stages, alternative terms are used for the laser beams tailored from the light generated from the ECDLs. To avoid confusion, we add wording serving the purpose of these tailored light beams; for instance, the cooling and repumper beams designated for MOT operations will be specifically referred to as ‘*cooling-MOT*’ and ‘*repumper-MOT*’, respectively. Using such terminology also allows the reader to trace the beams back to their initial generation and stabilization scheme.

The output from the ECDLs is used to seed the tapered amplifiers (TAs) in a MOPA (Master Oscillator Power Amplifier) configuration. The ECDLs and TAs are integrated within the same module, streamlining the process and eliminating the need for external alignment of the seeding light. In a MOPA configuration, a master oscillator—in this case, the ECDLs—generates a low power beam that is then amplified by the tapered amplifiers. These amplifiers are capable of producing more than 700 mW of power each. The amplified light from the two amplifiers is then coupled into polarization-maintaining fibers and distributed to an optical table located in proximity to the single-chamber vacuum system.

The frequency stabilization of the ECDL outputs is achieved through Doppler-free saturated absorption spectroscopy, facilitated by the commercially available ‘*CoSy*’ module from *TEM-Messtechnik*. The output signal from this module is then processed via a lock-in detection scheme, where the laser control current undergoes modulation to influence the laser frequency. Notably, both lasers incorporate current modulation at frequencies of 41.12 kHz for the repumper and 45.96 kHz for the cooling laser. This modulation is crucial for generating an error signal, effectively functioning as a lock-in signal, and is a widely employed technique in signal processing methods.

The error signal is obtained via the modulation of the laser frequency, which translates into an amplitude modulation when the light’s frequency is modulated around a spectral feature. This amplitude modulation is then demodulated by mixing it with an RF signal used in the modulation of the frequency of the light. Subsequently, this demodulated and synchronized signal yields a DC signal proportional to the derivative of the signal obtained from Doppler-free saturated absorption spectroscopy. Thus providing an error signal for laser stabilization. The stabilization via this generated error signal ensures that the frequency drift, both short and long-term, is much smaller than the linewidth of the cooling and repumping transition (approximately 5.2 MHz [45]), a crucial requirement in ultracold experiments. However, the modulation of the laser frequency leads to a broadening of the linewidth in these lasers. This broadening is quantified by obtaining the beat note signal between an optical frequency comb and the cesium lasers, as illustrated in Figure 4.4 for the cooling laser, with its linewidth approximately $\Delta\nu_{\text{FWHM}} \simeq 4$ MHz.

The laser frequency is stabilized by tuning the current fed to the ECDLs using a proportional-integral-derivative (PID) loop. Specifically, the repumper light is stabilized to the $6s^2S_{1/2}(F = 3) \rightarrow 6p^2P_{3/2}(F' = 3)$ transition. The selected frequency for the stabilization of the cooling laser differs slightly. The seeding light of the cooling laser undergoes a frequency alteration of -69 MHz using an AOM (minus first order) before being sent to the Doppler-free saturated absorption spectroscopy module. The obtained error signal with the frequency-altered light was used for stabilizing the laser to the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 4)$. In Figure 4.5, the transitions targeted for stabilizing the laser frequencies of the repumper and cooling lasers are depicted alongside their respective error signals.

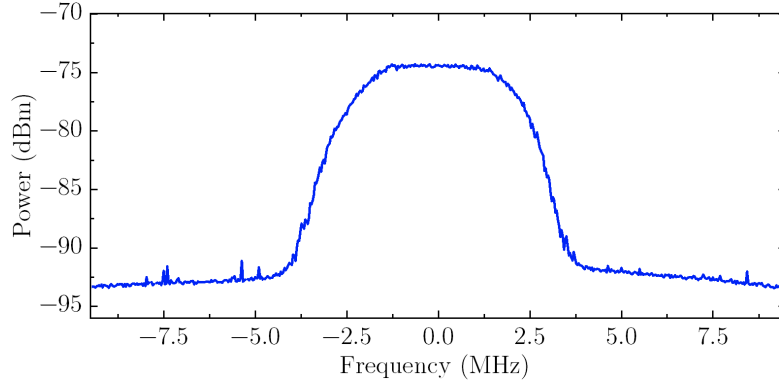


Figure 4.4: The frequency spectrum of the cesium cooling laser when its stabilization is active. Modulation of the laser frequency to obtain the error signal results in the broadening of the linewidth. The frequency profile is obtained via a beat note (a signal representing the difference in optical frequencies) between an optical frequency comb [35] and the cesium cooling laser. The central frequency is adjusted to zero for visual clarity, whereas it measured 67.8 MHz in practice.

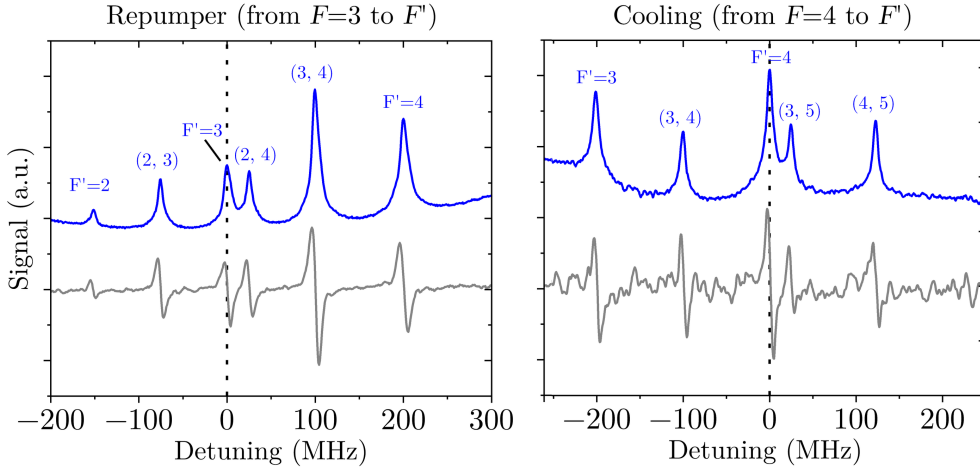


Figure 4.5: Doppler-free saturated absorption spectroscopy spectra on the D_2 -line of cesium (blue). The lower levels are $6s^2S_{1/2}$, $F = 3$ (left), and $F = 4$ (right), whereas the upper levels belong to the $6p^2P_{3/2}$ manifold. All transitions allowed by selection rules can be clearly resolved, including crossover resonances (marked in brackets). The error signal for frequency stabilization, shown in grey, is proportional to the derivative of the signal displaying spectral features.

As a result, the amplified cooling light possesses a frequency of +69 MHz from this transition. The necessity to alter the frequency of the seeding cooling light can be explained by considering the timeline of the laser system construction. During the construction of this system, our laboratory only had access to AOMs designed for optimal usage with driving frequencies of 80 MHz and 110 MHz. As a result, the frequency alteration was necessary with the requirement to generate light frequencies that can be subsequently tailored for further experimental methods for cooling and imaging of cesium atoms (as illustrated in Figure 4.8, placed at the end of the section).

The stabilized cooling and repumper laser beams are directed to another optical table where the frequency, intensity, and polarization of the prepared light beams are adjusted to meet the specific requirements of the methods being performed. The prepared light frequencies are shown in Figure 4.8 at the end of this section. Currently, our setup has the capability to maintain an MOT, perform GMC [122, 123], selectively populate atoms in the ground state energy levels $F = 3$ and $F = 4$, and produce spin-polarized samples in the spin states $F = 3$, $M_F = 3$ or $F = 3$, $M_F = -3$ states.

The preparation of these beams relies on two main optical setups (cooling-path and repumper-path as illustrated in Figure 4.6), where in one, the cooling laser light is split into different paths for the generation of cooling-MOT, cooling-GMC, and absorption imaging beams. On the other, the repumper light is divided into paths to prepare repumper-MOT and spin-polarization beams.

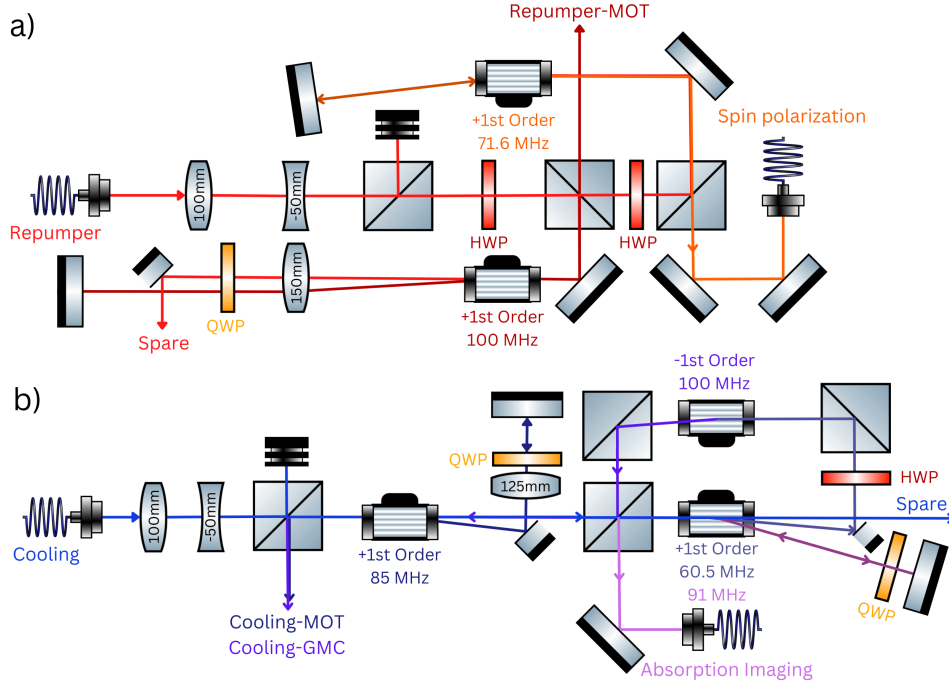


Figure 4.6: Optical setup layouts for: a) preparation of the repumper-MOT and the spin polarization, b) preparation of cooling-MOT, gray molasses cooling (GMC), and absorption imaging. Optical beam paths are color-coded to correspond with the driving frequencies of the AOMs, which are also color-coded. QWP: Quarter-waveplate. HWP: Half-waveplate. *The optical components depicted in this figure are licensed under Ref. [121].*

Based on the principles underlying our experimental methods, we designed this setup with consideration for beam generation, allowing for potential simultaneous use of beams, if needed. For instance, when the cooling-MOT beam operates at its maximum optical power, it may limit the preparation of the absorption imaging beam to the desired power level. However, it is essential to note that these two beams serve distinct purposes and typically do not need to be prepared simultaneously.

To begin with, the preparation of MOT beams involves dual-pass AOM configurations, where the desired frequencies are achieved by setting the driving frequency of the AOM to 100 MHz for the repumper-MOT and 85 MHz for the cooling-MOT. The dual-pass configuration is essential not only to alter the frequency of the light by twice the driving frequency of the AOM, but also to enable frequency tuning without changing the position of the beam. Given the significant length of the optical path from the AOMs to the experimental chamber (~ 2.5 m), even a minor deviation in the angle of the outgoing light from the AOM can lead to the MOT beams being clipped off, as they may partially exit the aperture of optical elements along the path.

In the same optical setup, the absorption imaging beam is created by allowing the cooling beam to pass through the first AOM (zeroth order), as shown in [Figure 4.6b](#). This beam goes through a second AOM in the setup, where the diffracted first-order beam from this second AOM is utilized in a dual-pass configuration with a driving frequency of 91 MHz. After passing through the second AOM and a quarter-waveplate (QWP) twice, the beam is extracted through a polarizing beam splitter (PBS) and coupled into a polarization-maintaining single-mode fiber. This fiber is employed in the distribution of the beam to the absorption imaging setup, details of which are explained in [Section 4.4.1](#).

Simultaneously, the second AOM in the cooling path generates a beam for gray molasses cooling (GMC). GMC is a sub-Doppler cooling technique that employs a blue-detuned cooling laser to induce atomic transitions of $F \rightarrow F' = F$ or $F \rightarrow F' = F - 1$ [48, 53], as briefly discussed earlier in [Section 2.2](#). In our setup, the necessary blue-detuned beam is generated by driving the second AOM in the cooling path with 60.5 MHz; its first-order beam is then redirected to a third AOM with a driving frequency of 100 MHz. The resulting -1st order beam from the third AOM is overlapped with the cooling-MOT beam's path, passing through the MOT beam distribution setup (as shown in [Figure 4.7](#)) before reaching the sample. Consequently, a 29.5 MHz (5.54Γ) blue-detuned beam from the transition $F = 4 \rightarrow F' = 4$ is prepared.

Moreover, the spin-polarization beam is produced in the repumper path (refer to [Figure 4.6a](#)) using another AOM in a double-pass configuration with a driving frequency of 71.6 MHz. The generated beam is then coupled into a polarization-maintaining optical fiber for transmission through the sample. More detailed explanation on the usage of this generated beam is provided in [Section 5.3](#).

The generated cooling-MOT (and cooling-GMC beam sharing the same optical path) and repumper-MOT beams are directed into three distinct optical paths named MOT-1, MOT-2, and MOT-3 (refer to [Figure 4.7a](#)). Initially, a polarizing beam splitter is used to overlap these two beams, and their initial polarization orientations are adjusted using a half-waveplate for desired power splitting ratios.

For the cooling beams, one-third of the input beam power is directed to the MOT-1 path, and for the repumper-MOT, the distribution is organized such that MOT-2 and MOT-3 carry half of the input power. Subsequently, a second PBS is employed to equally split the beams, ensuring uniform power distribution among all three MOT beams for cooling beams and equal distribution for repumper-MOT in the paths MOT-2 and MOT-3. The second PBS also ensures that the polarization of the cooling and repumping beams is identical after the splitting. After the last PBS, which directs the beams to each MOT beam path, we use a quarter-waveplate (QWP) and a half-waveplate (HWP) to ensure that the polarization becomes circular before reaching the experimental chamber. The need for a QWP and an HWP is particularly due to the polarization

disturbances caused by the mirrors and the dichroic mirrors that overlap the potassium and cesium light paths. The QWP alone is insufficient to achieve the desired circular polarization (σ^- for MOT-1 and σ^+ for MOT-2 and MOT-3) in the presence of the mentioned optical elements on the path.

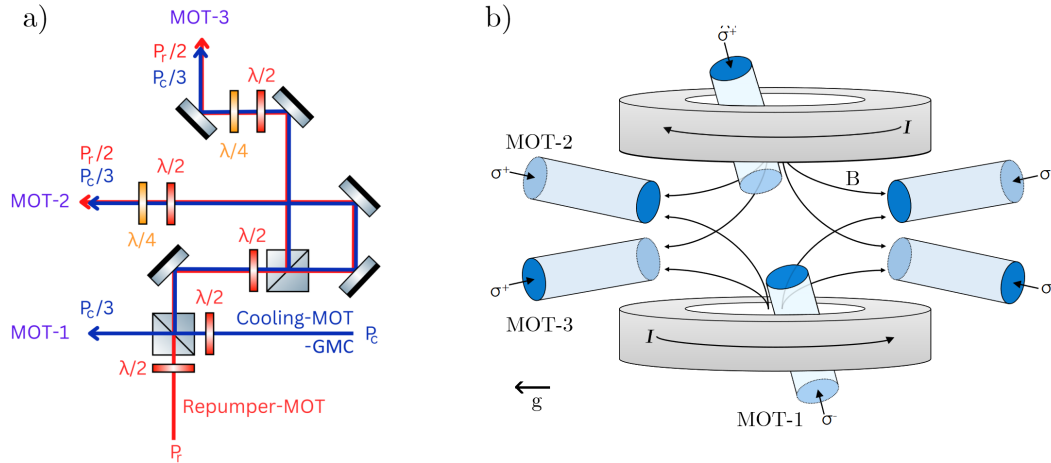


Figure 4.7: a) Layout of the optical power distribution setup for repumper MOT, cooling MOT, and cooling-GMC beams. P_r and P_c refer to the input laser power of cooling-MOT (or cooling-GMC) and repumper-MOT beams individually. b) Illustration of the laser beams and the electromagnetic coils for the MOT of KCs single-chamber. The glass cell and the additional coils are not shown for simplicity. The optical components depicted in this figure are licensed under Ref. [121].

Beams that are directed to the MOT paths are then enlarged by a factor of three via a set of lenses, resulting in around 27 mm of beam size. Paths MOT-2 and MOT-3 are then directed to the experimental chamber with 50° and 130° angle to the z -axis, in which the optical axes of these beams are in the y - z plane. MOT-1 path has the angle of 15° with respect to the remaining axis (x) in the tilted x - z plane. In Figure 4.7b, an illustration depicting the arrangement of laser beams and electromagnetic coils for utilizing the MOT is presented. By using an identical set of lenses, the beam sizes are reduced to fit into the aperture of available quarter-waveplates. Finally, the beams were retro-reflected with mirrors, and a double-passed quarter-waveplate was utilized to tune the polarization to the desired orientation.

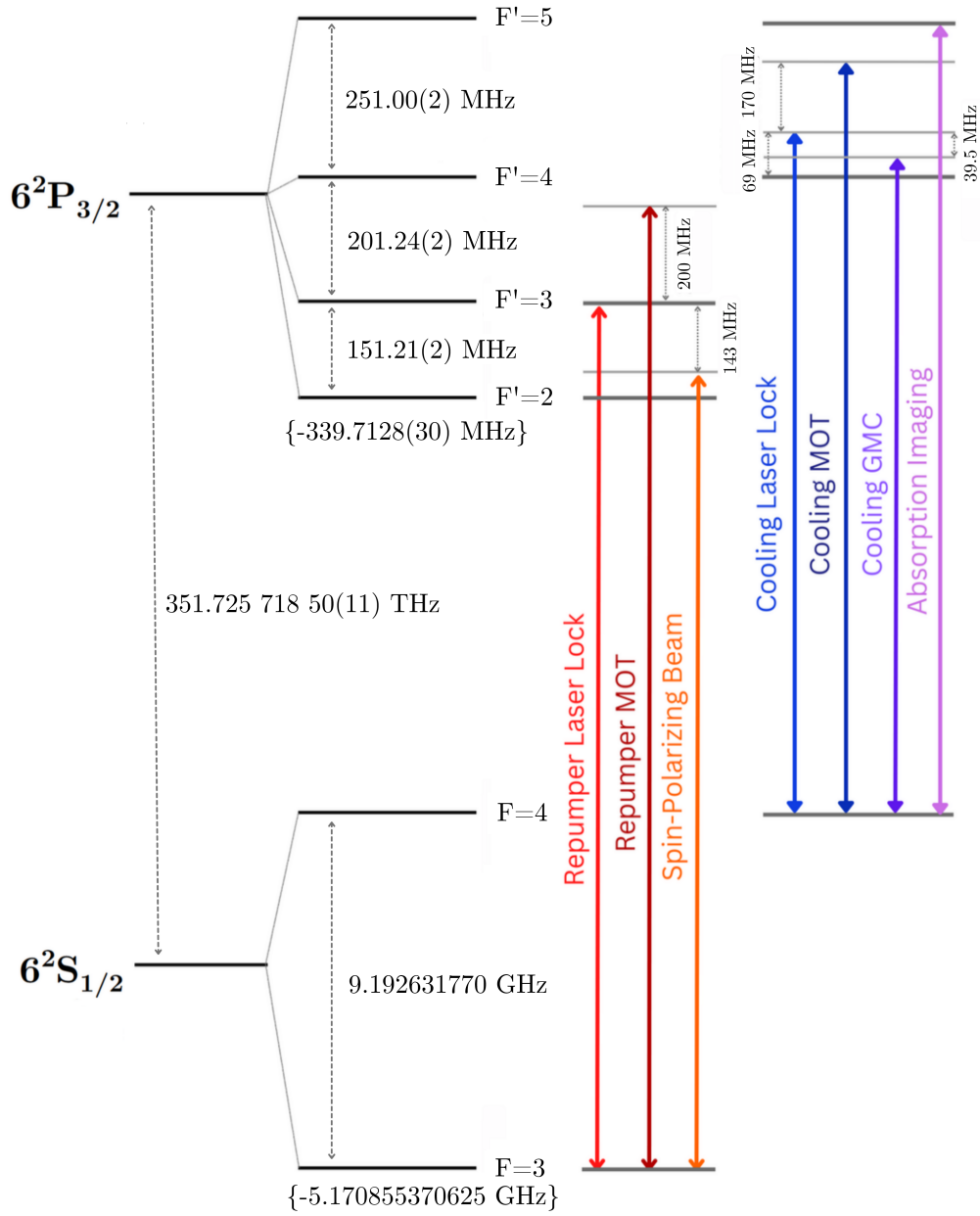


Figure 4.8: Illustration showing the Cesium D₂ hyperfine transitions (hyperfine splittings are taken from Ref. [45]). The corresponding light frequencies for various cooling and probing methods are also shown. The repumper laser beam serves the dual purpose of preparing repumper-MOT beams and the spin-polarizing beam. In contrast, the cooling laser beam is employed for cooling-MOT, cooling GMC, and absorption imaging beams. The definitions of the beams and their roles can be found in this section. Frequencies denoted in brackets below $F = 3$ and F' represent the difference between these levels and their corresponding fine-structure levels.

4.2.2 Dipole trapping laser

Optical dipole traps are used for ultracold experiments to create an optical field that allows precise three-dimensional confinement of the atoms and molecules of interest [124, 125, 126]. This phenomenon relies on the theory of optical dipole force, which is enabled by the interaction of the induced atomic dipole moment with the electric field gradient created by a laser beam [127]. Briefly, the dipole moment of the atom arises due to the displacement of the electron cloud from the atomic nucleus in response to the electric field of the laser beam. This displacement leads to a separation of charge, resulting in an induced dipole moment. The optical force acting on the atom is proportional to the gradient of the electric field generated by the laser beam. The intensity gradient, which is the spatial variation of the laser beam's intensity, determines the trapping potential's depth and shape. Thus, a focused beam can create a potential with a minimum (maximum) at the focal point if the frequency of the laser is red-detuned (blue-detuned) concerning atomic transitions. Figure 4.9a illustrates the red-detuned trapping potential generated with a Gaussian beam, while Figure 4.9b demonstrates the effect of gravity on cesium atoms within the same trapping potential. Cesium atoms, being relatively heavy compared to other atomic species, induce a higher disturbance on the optical dipole traps, significantly reducing the effective trap depth.

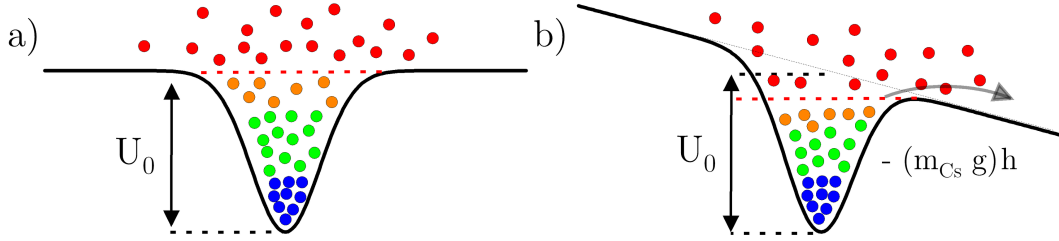


Figure 4.9: Potential energy curves of a red-detuned optical dipole trap at the focus showing a) one-dimensional cross-section perpendicular to the direction of gravitational force, b) along the direction of the gravitational force. The arrow illustrates how the addition of gravity tilts the Gaussian potential, allowing atoms to move beyond the confines of the potential and exit the trap. The atoms are color-coded with respect to their temperatures, blue being the coldest and red being the hottest. The U_0 represents the trap depth, while $-(m_{Cs}g)h$ corresponds to the gravitational potential acting on cesium atoms. The grey arrow represents the potential spillage of atoms from the trap due to the acting gravitational force.

The dipole trapping potential can be spatially tuned by adjusting the profile of the trapping laser beam. For instance, a steep intensity gradient can be achieved through a tightly focused beam, allowing for a larger trapping depth with a smaller trapping volume. Conversely, while maintaining the same beam power, a less tightly focused beam results in an expanded trapping volume with smaller trapping frequencies and reduced trap depth. This also enables the formation of complex dipole potentials for confining atoms within spatially defined intensity profiles [128, 129]. In the course of this thesis, we undertook the task of shaping spatial beam profiles using a digital micro-mirror array for the utilization of custom-made intensity profiles. However, it is worth noting that the specific topic and outcomes are not directly related to the context of photoassociation and were not included in the thesis. The dipole interaction potential generated via an optical trapping beam can be described as

$$U_{dip}(r) = -\frac{3\pi c^2}{2\omega_0^3} \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega} \right) I(r), \quad (4.1)$$

and the scattering rate by the presence of this dipole beam is

$$\Gamma_{sc}(\mathbf{r}) = \frac{3\pi c^2}{2\hbar\omega_0^3} \left(\frac{\omega}{\omega_0} \right)^3 \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega} \right)^2 I(\mathbf{r}). \quad (4.2)$$

In the given context, c represents the speed of light, ω is the driving frequency corresponding to the dipole trapping laser frequency, and ω_0 signifies the transition frequency from the ground state to the excited state of interest. The damping rate, denoted by Γ , reflects the rate at which the excited state spontaneously decays. $I(r)$ represents the intensity profile of the trapping beam(s). It can be expressed in terms of the dipole matrix element between the ground and excited states:

$$\Gamma = \frac{\omega_0^3}{3\pi\epsilon_0\hbar c^3} |\langle e|\mu|g\rangle|^2, \quad (4.3)$$

where, μ represents the transition matrix element.

If two atomic transitions, as in the case of cesium atoms, D_1 and D_2 lines, are closely related to the frequency of the dipole trap laser, their influence cannot be neglected. In such cases, [Equation 4.1](#) can be appropriately reformulated as:

$$U_0 = \frac{3c^2 \cdot P}{w^2} \left(\frac{\Gamma_{D1}}{\omega_{D1}^3} \left(\frac{1}{\omega_{D1} - \omega_0} + \frac{1}{\omega_{D1} + \omega_0} \right) + 2 \frac{\Gamma_{D2}}{\omega_{D2}^3} \left(\frac{1}{\omega_{D2} - \omega_0} + \frac{1}{\omega_{D2} + \omega_0} \right) \right). \quad (4.4)$$

Here, P is the power of a Gaussian trapping beam and w is the width of the trapping beam. The factor of two before the second term in the bracket represents the relevant line strength factors favoring the D_2 line. The trap depth of an optical dipole trap with respect to the wavelength of the trapping laser is given in [Figure 4.10](#) for cesium atoms.

Our experimental setup includes the implementation of a dipole trap, driven by the recognition that optical dipole traps offer significant enhancements. The benefits of having such a system are briefly explained below, with a particular focus being on photoassociation measurements.

Optical dipole traps play a crucial role in further cooling samples, which are beneficial not only for photoassociation experiments but also for all ultracold experiments. Specifically, we have employed ‘evaporative’ cooling [\[52, 130, 131, 132\]](#) and are in the process of implementing degenerate Raman sideband cooling [\[51, 133\]](#), which is still under development in our KCs apparatus and was not utilized within the timeframe of this thesis.

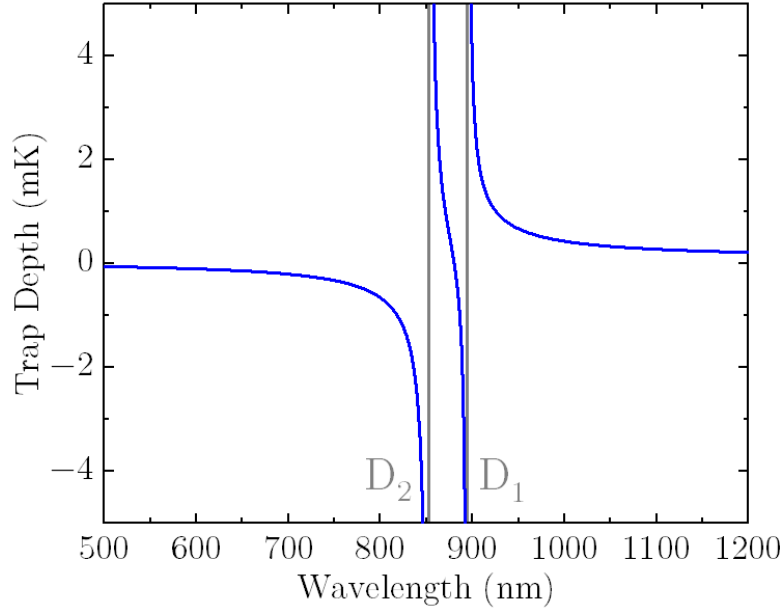


Figure 4.10: Trap depth of an optical dipole trap for varying wavelengths of the trapping beam used for the confinement of cesium atoms. The trap depth in the vicinity of both atomic transitions (D_1 and D_2) is heavily influenced by both transitions.

The evaporative cooling method is commonly employed to cool an atomic sample to a lower temperature by allowing higher-energy particles to escape or evaporate from the system. In an optical dipole trap, evaporative cooling is achieved by gradually decreasing the intensity of the lasers over time, thereby reducing the trap depth. As the intensity decreases, particles in the sample with the higher energies escape more easily, leaving behind a colder and denser sample of atoms. Despite achieving denser and colder samples through the evaporation of our ultracold atomic ensembles, the overall atom count was significantly reduced due to atom loss resulting from collisions between the ultracold atoms and the hot background gas, as expected from the design of the single chamber apparatus. The evaporative cooling scheme was infrequently employed in the photoassociation measurements due to the constraints of the short lifetime of our optical trap and the required timings for conducting evaporative cooling, exerting a significant reduction of the resulting number of atoms before the start of the photoassociation stage.

Additionally, when atoms are confined in an optical dipole trap, a uniform magnetic field can be applied to manipulate the collisional properties of an atomic sample by tuning the scattering length of the trapped samples [134] — a capability not possible in a MOT. The limitation of using uniform external magnetic fields in MOTs is that it changes the location of the magnetic field gradient minima, leading to a shift in the position of the trapping center. Depending on the amplitude of the applied uniform field, the trapping center may move the ultracold sample out of the apertures of the optical beams, making it challenging and often impossible to utilize the cooling process efficiently.

In the context of this thesis, external uniform magnetic fields are utilized for the manipulation of the molecule formation rate through a technique known as Feshbach-optimized photoassociation

[27, 135, 136] (also detailed in Section 5.4). This capability enhances experimental control over the photoassociation process and increases the likelihood of forming excited molecular levels when exposed to laser resonant to a photoassociation transition in the vicinity of a Feshbach resonance.

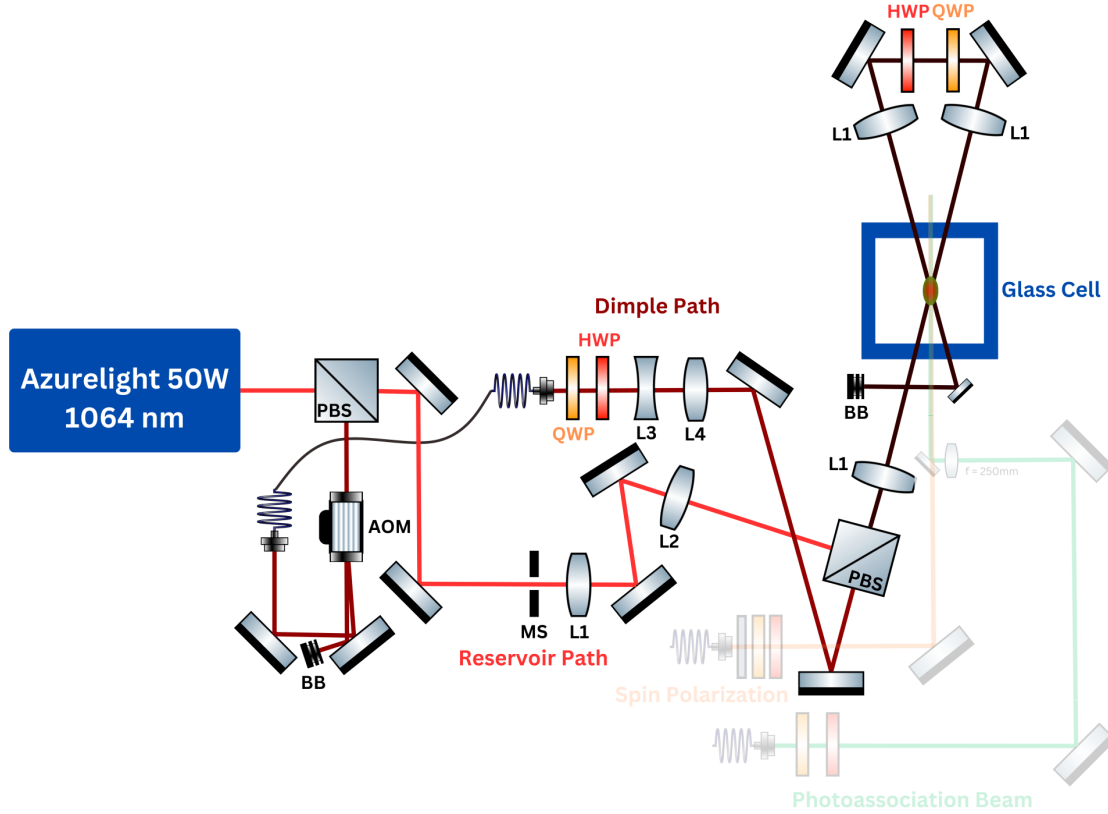


Figure 4.11: Optical schematic depicting the setup for optical dipole trapping. The paths of the dimple beam and the reservoir beam converge at the polarizing beamsplitter (PBS) with a high power damage threshold positioned near the glass cell. Key components include the Quarter-waveplate (QWP), Half-waveplate (HWP), Mechanical shutter (MS), and Beam block (BB). *The optical components depicted in this figure are licensed under Ref. [121].*

As described above, the implementation of the optical dipole trap system in our experimental apparatus provides significant contributions to our measurements. Our optical dipole trap is utilized by a 50 W commercially available laser amplifier ‘Azurelight 50W’ manufactured by Azurlight Systems. This laser amplifier is seeded with a narrow linewidth (<5 KHz as specified by the manufacturer) 1064.46 nm fiber laser (*NP Photonics RFLM-25-1-1064.46-1*) to generate a far-red-detuned laser beam suitable for both potassium and cesium atoms. Far-detuned optical dipole potentials are employed to reduce the scattering rate, which can otherwise result in the undesired loss of a significant amount of atoms from the trap. The dipole potential scales inversely with detuning as I/Δ (Equation 4.1), following the relationship I/Δ^2 for the scattering rate (Equation 4.2), where I is intensity and Δ is the detuning from the closest atomic transition. Far-detuned high-intensity beams are typically utilized to minimize the scattering rate, ensuring

a lower scattering rate and the intended potential depth.

The laser generating the dipole trapping beam operates in a single longitudinal and transversal mode. The resulting output beam is a single-mode (TEM_{00}) with the beam-quality factor being $M^2 \leq 1.1$ and employed for the optical dipole trapping process. The amplified output from the device is a collimated linearly polarized beam with a diameter of 1.0 ± 0.2 mm. The polarization is precisely vertically oriented, with a polarization ratio exceeding 300:1. Shortly after exiting the laser head, the beam is split into two paths on a polarizing beam splitting cube into two paths, called here the ‘*dimple*’ and the ‘*reservoir*’ paths (shown in Figure 4.11).

The reservoir beam facilitates the efficient transfer of atoms from optical molasses to the optical dipole trap [52] due to its larger volume than the dimple. At the initial stages of the experiment, a larger size of the reservoir beam is a crucial factor in minimizing the density of atoms in the trap. This reduction is necessary to address three-body losses, a persistent challenge in cesium experiments [137]. Subsequently, once the sample is loaded into the optical dipole trap formed by the reservoir beam, the dimple beam is then ramped up to confine the atoms loaded into the reservoir to create a denser sample (see Figure 4.12).

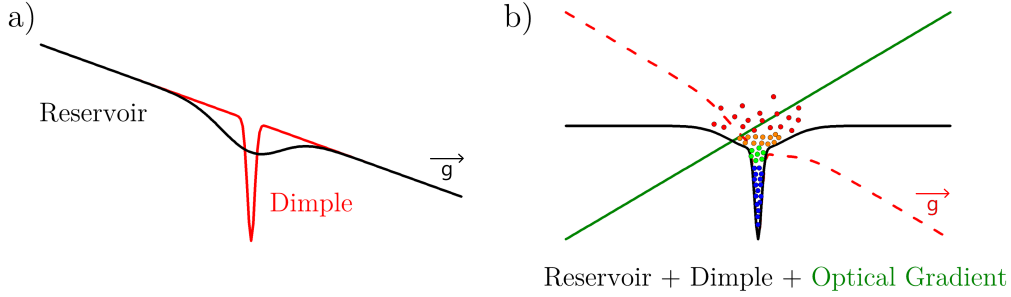


Figure 4.12: Potential energy curves of a red-detuned optical dipole trap with reservoir and dimple beams at the focus, showing: a) A one-dimensional cross-section along the direction of gravity, distinguishing the potential energy generated via the reservoir (black) and the dimple beam (red). b) The resulting potential energy curve when both the reservoir and dimple beams are active as a sequential part of loading the atoms from the reservoir to the dimple. The red dashed line represents the case where the gravitational effect is present. Disturbance of the gravity on the optical dipole potential can be eliminated by exerting force on the atoms in the opposite direction of gravity. This can be done in several ways, such as using magnetic field levitation [134, 138] and optical levitation [139]. The green line represents the necessary potential energy curve to compensate for the gravitational tilt, and the black line represents the case where gravity is compensated for or non-existent. The atoms are color-coded based on their temperatures, with blue indicating the coldest and red indicating the hottest.

Additionally, the atoms with higher kinetic energies than the dimple’s trap depth alone will remain confined within the reservoir, forming a pool of atoms to be loaded back into the dimple. In fact, this is why it is referred to as a ‘*reservoir*’ trap.

The laser beam power splitting ratio between the reservoir and dimple beam paths is initially adjusted using a half-waveplate mounted on a motorized rotational stage. The dimple path includes an AOM for robust intensity control, facilitating smooth sample loading from the reser-

voir and enabling evaporation. The light diffracted by an AOM typically disrupts the beam's output profile, which in turn also disturbs the generated trapping potential. Therefore, the light diffracted by the AOM is coupled into a polarization-maintaining single-mode fiber to ensure that the outgoing light is free from spatial impurities. Apart from their differences in spatial beam sizes, the two trapping beams have orthogonal polarization orientations.

To achieve the desired beam sizes for the dimple and reservoir beams, we use a set of lenses in each path. A beam waist of $250\ \mu\text{m}$ is chosen for the reservoir and $45\ \mu\text{m}$ for the dimple beam at the focus. Subsequently, the beams are spatially overlapped through the utilization of a polarizing beamsplitter, and both are focused at the center of the experimental chamber, as illustrated in [Figure 4.11](#). The overlapping of these beams is essential to ensure the optimal transfer of atoms between them. Finally, the beams are collimated at the opposite end of the chamber and redirected back into the experimental chamber. They form an angle of 40° between the returning beam and the initially passed beam, resulting in a crossing angle of 140° , with due consideration given to their wave-vectors (see [Figure 4.13](#)).

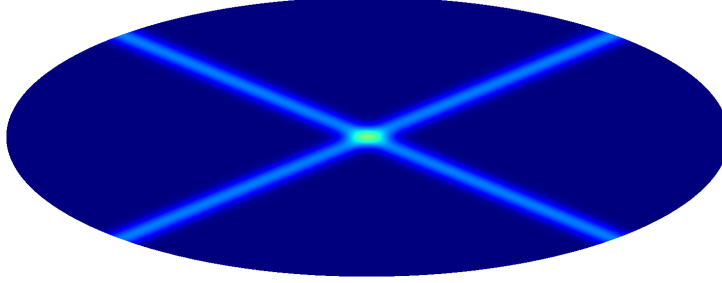


Figure 4.13: Top-down view of the numerically simulated crossed dipole trap beams (reservoir), with a 140° crossing angle between the incoming and outgoing trapping beams. Visualization is based on the value of the local intensity.

In the returning path, a half-waveplate and a quarter-waveplate are employed to control the polarization of the beam before it is refocused on the sample. Notably, if the polarization of the returning beam is chosen to be orthogonal to the first passing beam, an optical dipole trap is realized. On the other hand, if the waveplates are tuned so that the polarization is aligned in the same linear orientation as the incoming beam, an optical lattice is formed to confine the atoms.

It is worth noting that the presence of focused high-power beams influences the hyperfine energy levels of the atoms through the introduced AC Stark shift [\[140\]](#). The Stark effect refers to the shifting and splitting of the spectral lines of atoms and molecules caused by an external electric field. These spectral shifts are undesired when the intention is to carry out other cooling methods or obtain imaging readouts. We have installed a controllable mechanical shutter in the reservoir beam path, blocking the optical dipole trapping beam during the initial cooling phases (typically $t_c < 8\ \text{s}$) and in the imaging phase (typically $t_i < 50\ \text{ms}$). Similarly, the AOM in the dimple path is turned off during these stages, directing the beam to a physical beam block.

For a better understanding of the employed optical trapping method, two important variables are numerically calculated: trap frequencies and trap depth. The trap frequencies for a crossed dipole trap can be determined using the following expressions [\[141, 142\]](#):

$$\omega_x(\theta) = \sqrt{\frac{4U_0}{m}} \sqrt{\frac{2\cos^2\theta}{w_0^2} + \frac{\sin^2\theta}{z_r^2}}, \quad (4.5)$$

$$\omega_y(\theta) = \sqrt{\frac{8U_0}{mw_0^2}}, \quad (4.6)$$

$$\omega_z(\theta) = \sqrt{\frac{4U_0}{m}} \sqrt{\frac{\cos^2\theta}{z_r^2} + \frac{2\sin^2\theta}{w_0^2}}, \quad (4.7)$$

where, U_0 is the depth of the trap for a single Gaussian beam, w_0 is the width of the beam at focus, and z_R is the Rayleigh range. In our case, the y direction is along the direction of gravity, θ is the angle between a single Gaussian beam and the z-axis, with 2θ being the angle between the crossed beams.

Similarly, when our trapping beams are used for generating a 1D lattice by having both dipole trapping beams to have the same polarization, the trap frequencies can be described as the following [127, 143, 144]:

$$\omega_x(\theta) = \sqrt{\frac{8\pi^2 U_0}{md^2}}, \quad (4.8)$$

$$\omega_y(\theta) = \sqrt{\frac{16U_0}{mw_0^2}}, \quad (4.9)$$

$$\omega_z(\theta) = \sqrt{\frac{16U_0}{mw_z^2}}, \quad (4.10)$$

where,

$$w_z(\theta) = \sqrt{\frac{1}{w_0^2 \sin^2(\theta)} + \frac{\cos^2(\theta)}{2z_r^2}}. \quad (4.11)$$

Here, d represents the period of the 1D lattice, and it can be expressed as $d = \lambda_{ODT}/(2\sin\theta)$. In our specific case, with θ set at 70° , the calculated value for d is 566 nm.

Utilizing the equations mentioned above (Equations 4.5-4.11), one can calculate the trap depth and trap frequencies for both the reservoir and dimple confinements for the ^{133}Cs atoms (refer to Table 4.1). The trap configuration for the reservoir and dimple can also be determined based on their respective confinement types, whether they function as dipole traps or optical lattices. These parameters are calculated using typical beam powers: 40 W for the reservoir beam and

Table 4.1: Trap depth and trap frequencies obtained via Equations 4.5-4.11 with the reservoir beam power of 40 W and dimple beam power 8 W.

	Reservoir		Dimple	
	Dipole	Lattice	Dipole	Lattice
Trap Depth	316.33 μ K	1.26 mK	1.95 mK	7.81 mK
ω_x	86.63 Hz	351 kHz	1.19 kHz	873.30 kHz
ω_y	253.30 Hz	358.22 Hz	3.49 kHz	4.94 kHz
ω_z	238.02 Hz	336.61 Hz	3.28 kHz	4.64 kHz

8 W for the dimple beam. Consequently, in Figure 4.14, the trap depth and frequencies versus the dipole trapping laser power can be seen.

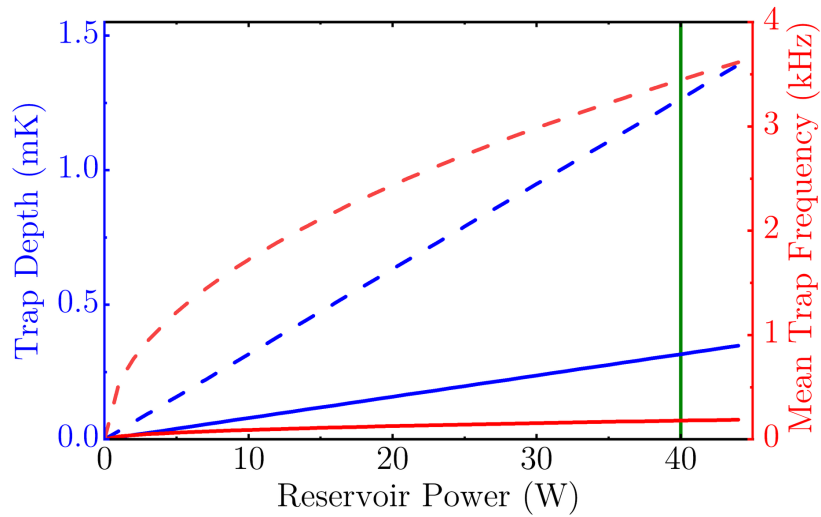


Figure 4.14: The relationship between the optical dipole trap depth and the optical lattice trap depth is given as a function of the reservoir beam power when the crossing beams are at an angle of 140° . Continuous lines represent the dipole trap configuration. Dashed lines represent the optical lattice configuration. The mean trap frequencies are provided for both configurations. The green line indicates the typical reservoir beam power selected for photoassociation experiments.

It is important to note that the lattice configuration has multiple pancake-shaped potentials. The formulations and calculations given in this part are only focused on the central trapping site. If necessary, it is advisable to analyze them individually, as they exhibit different trap depth values in terms of the dipole potential they generate.

The experimental sequence, which consists of loading the dipole trap and operating it for various controlling processes, is given later in this thesis, namely in Section 5.2.

4.3 Magnetic field sources

The electromagnetic coil system introduced in this section is designed to serve multiple purposes. Primarily, it facilitates experiments involving the cooling of atoms to ultracold temperatures and enables precise control over their collisional properties by adjusting external magnetic fields. Additionally, it defines the quantization axis at various stages of the experimental sequence. This section not only presents the setup of the coil system but also offers insights into the process of characterizing these magnetic field sources. The characterization is a crucial step to ensure the reliability and accuracy of the magnetic fields used in experiments, particularly those involving ultracold atoms. The section covers the details of how these coils are assessed and calibrated to meet the specific requirements of our research.

4.3.1 Electromagnetic coils

In our single-chamber KCs apparatus, we employ five sets of electromagnetic coils arranged in a cage-like structure around the experimental cell to generate the necessary magnetic fields for cooling and manipulating ultracold atoms. This arrangement compensates for the existing stray field by providing the ability to tune the magnetic field in all three dimensions. Additionally, the setup enables the generation of magnetic field gradients for magnetic trapping and uniform high fields in a single axis, as required for various ultracold experiments. The cage-like structure offers convenient optical access to the scientific cell, as illustrated in [Figure 4.15](#). These electromagnetic coil sets serve distinct purposes, each specifically designed for their intended function. To be more specific:

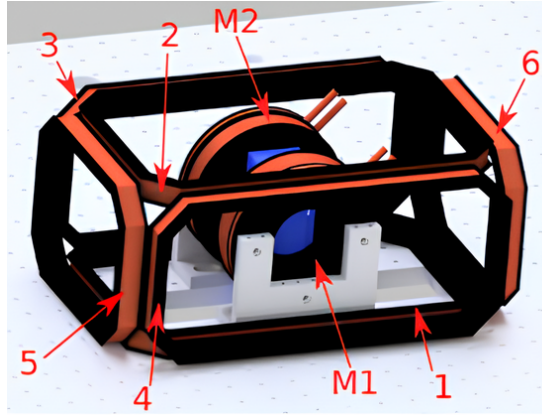


Figure 4.15: The rendered image of the magnetic coil system installed in the single-chamber experimental apparatus for K-Cs. M1 and M2 consist of two sets of electromagnetic coils: MOT and Feshbach. The numbered coil sets 1-3, 2-4, and 5-6 are compensation coils. The figure is taken from Ref. [120].

MOT Coils: The inner coils of each set (designated M1 and M2 in [Figure 4.15](#)) are designed to generate a field for a magneto-optical trap (MOT). These two coils operate with currents flowing in opposite directions to create the quadrupole field necessary for the MOT, a configuration known as an anti-Helmholtz configuration.

Feshbach Coils: The second set of coils (designated as the outer set of the M1 and M2 coils in [Figure 4.15](#)) is utilized to create a uniform magnetic field, enabling the tuning of atomic interactions through Feshbach resonances. These coils are connected in a Helmholtz configuration, with currents flowing in the same direction in both coils. The uniform magnetic field it produces can reach up to 300 G without requiring additional cooling methods. Furthermore, it can be further increased to 450 G when water cooling is activated, ensuring that the temperature of the coils remains within acceptable limits.

Compensation Coils: Isolating the system from external factors is crucial. While atoms in the vacuum cell are well isolated from most environmental factors, they can still sense external parasitic magnetic fields. Time-independent external magnetic fields are compensated for during experiments using the compensation coils. The set of rectangular compensation coils comprises three pairs (coils numbered from 1 to 6 in [Figure 4.15](#)) arranged in a Helmholtz configuration, enabling the tuning of the magnetic field in all three spatial directions. Simultaneously, when used in conjunction with the MOT coils, compensation coils can alter the position of cold atoms in the MOT by changing the center of the quadrupole field. This capability allows them to be utilized for magnetic levitation, as discussed in [Section 5.3](#).

I actively participated in the construction of these electromagnetic coils, a time-consuming process that involves meticulously positioning the wires within the assembly, ensuring precise alignment and spacing, and applying thermal paste to each layer of wire for every coil. This process is crucial for achieving optimal performance and reliability in the final product, as even minor errors in wire placement or thermal management can significantly impact the coil's efficiency and effectiveness.

4.3.2 Characterization of electromagnetic coils

Understanding the behavior of magnetic field sources before their integration into experimental setups is crucial. While simulations tend to match measurements closely, this alignment is often only evident after the magnetic field source has been characterized. Coils commonly used in ultracold experiments are home-built, employing ad hoc equipment to guide and secure copper wire. However, the resulting windings may not adhere to the idealized stack assumed in simulations. Additionally, mounting coils in the setup can introduce tilts, further deviating from the simulated arrangement.

In this context, the initial characterization of our electromagnetic coils installed on the KCs single chamber apparatus was done with a custom rig that utilizes a commercially available Hall Sensor (Hirst GM07) and a homemade linear translational stage where the sensor is attached [\[120\]](#). This setup needed development on its user-friendliness, and the characterization process was rather time-consuming. To address these concerns and improve our research efficiency for future projects, we developed an open-source magnetometer tailored for accurate magnetic field characterization in ultracold experiments [\[145\]](#) which is described in the following section. However, as the electromagnetic coils designed for the KCs single-chamber were already characterized, installed, and made operational, the developed open-source magnetometer was not employed in characterizing this particular setup, but it was used for characterizing electromagnetic coils made for other projects in our research group [\[146\]](#).

For the post-installation characterization of our installed electromagnetic coils, we employed

various methods to directly measure the influence of the generated magnetic fields on ultracold cesium atoms (conducted solely by myself) and potassium atoms (conducted by Mateusz Bocheński).

Automated magnetometer for pre-installation coil characterization

This subsection is based on our manuscript ‘Open-Source Magnetometer for Characterizing Magnetic Fields in Ultracold Experiments’ [145]. In this work, I was responsible for building the mechanical and electronic parts of the setup and developing the necessary code that enables data acquisition, processing, and communication with the user.

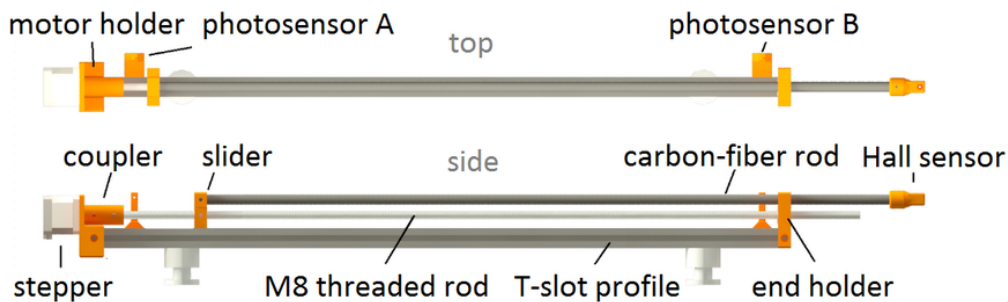


Figure 4.16: The arrangement of the automated magnetometer is visible from both a top-down and a side view. Components colored in orange have been 3D printed. A coupler connects the rotating stepper motor shaft to the threaded rod. Within the slider, an M8 nut is integrated to convert the rotational movement of the rod into the linear motion of the carbon fiber tube, which holds the Hall sensor. At the end, a holder with a ball bearing is employed to ensure the smooth rotation of the rod. Photosensors play a crucial role in determining the limits of motion, preventing collisions with the supporting structure in case information about the sensor’s current position is lost.

This section presents a novel, fully automated, open-source magnetometer tailored to characterize magnetic fields generated by various sources such as coils, permanent magnets, or parasitic elements. This developed open-source magnetometer relies on an Arduino Mega microcontroller and a three-axis Hall sensor with a ± 8 G per-axis measurement range, providing a root mean square (RMS) field readout below 0.3 mG. Notably, the sensor displacement during data acquisition is virtually limitless for practical purposes, making it particularly advantageous for characterizing large or extended coils such as Helmholtz cages or Zeeman slower [147, 148] (see Figure 4.16). All components required for building this system are cost-effective and widely available off-the-shelf items or can be created through 3D printing.

The newly developed magnetometer features a robust aluminum structure constructed with T-slot frames, allowing for convenient attachment of its components. The sensor achieves one-dimensional movement by being motorized through a stepper motor, traversing along the magnetic field source. The stepper motor shaft is connected to a threaded rod with a coupler. Smooth rotation of the threaded rod is assured by a ball bearing embedded in the other end of the rod. As the shaft of the stepper motor rotates the threaded rod, an M8 nut embedded in the slider

block enables the rotation of the threaded steel rod to be converted into the translational motion of the tube. The magnetometer's Hall sensor is attached via an adapter to this non-magnetic tube made of carbon fiber. The wires connecting the sensor to the electronics module of the magnetometer are guided through the inside of this carbon fiber tube. The assembly of the mechanical structures is fairly simple and described in the user's manual [149].

The electronic components of the automated magnetometer are controlled by an Arduino Mega single-board microcontroller (see Figure 4.17 for the schematics).

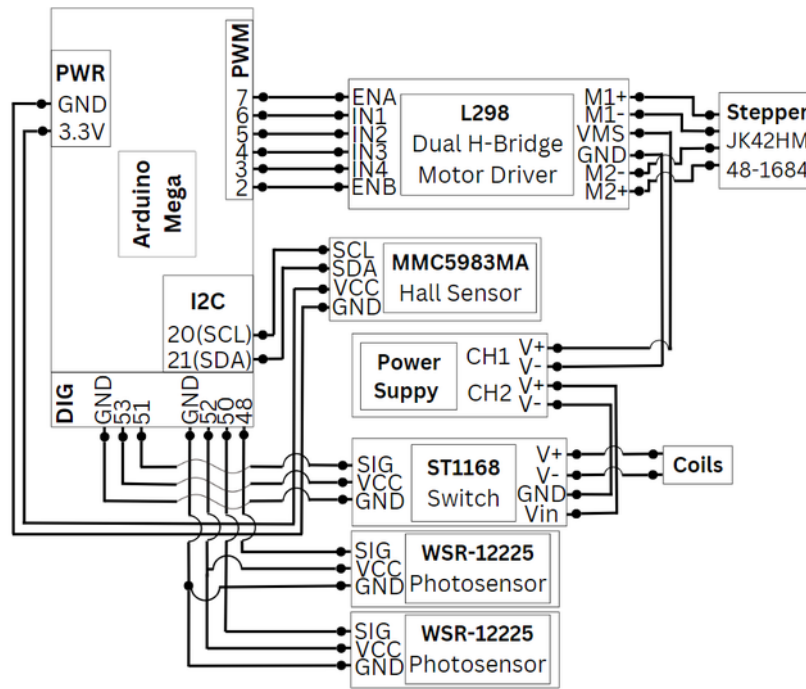


Figure 4.17: Schematic of the connections for the electronic components of the automated magnetometer. The Arduino Mega is controlled by a computer via a USB serial connection.

Typically, microcontrollers are programmed with specific firmware to carry out predefined tasks. However, making modifications to the setup, especially in open-source projects, can be challenging. To address this limitation, 'Telemetry-AIO' [150] was implemented into the microcontroller, a firmware built upon the generic Firmata protocol. Notably, this firmware not only supports all Firmata features but also incorporates stepper motor functionality and communication protocols like I²C, commonly utilized by various sensors. This protocol streamlines communication between the software running on a computer and the Arduino board. By uploading the Firmata firmware onto the microcontroller, APIs (application programming interfaces) are used to issue commands to execute desired tasks. All the essential functionalities required for controlling the magnetometer via APIs have been incorporated into a software developed using Python 3 software language. The magnetometer's operation is governed through a user-friendly graphical user interface (GUI) as shown in Figure 4.18, which oversees critical functions, including data acquisition and plotting. The GUI also incorporates supplementary features like data averaging, calibration of the Hall sensor displacement per step for the stepper motor, and real-time

monitoring of the present magnetic field.

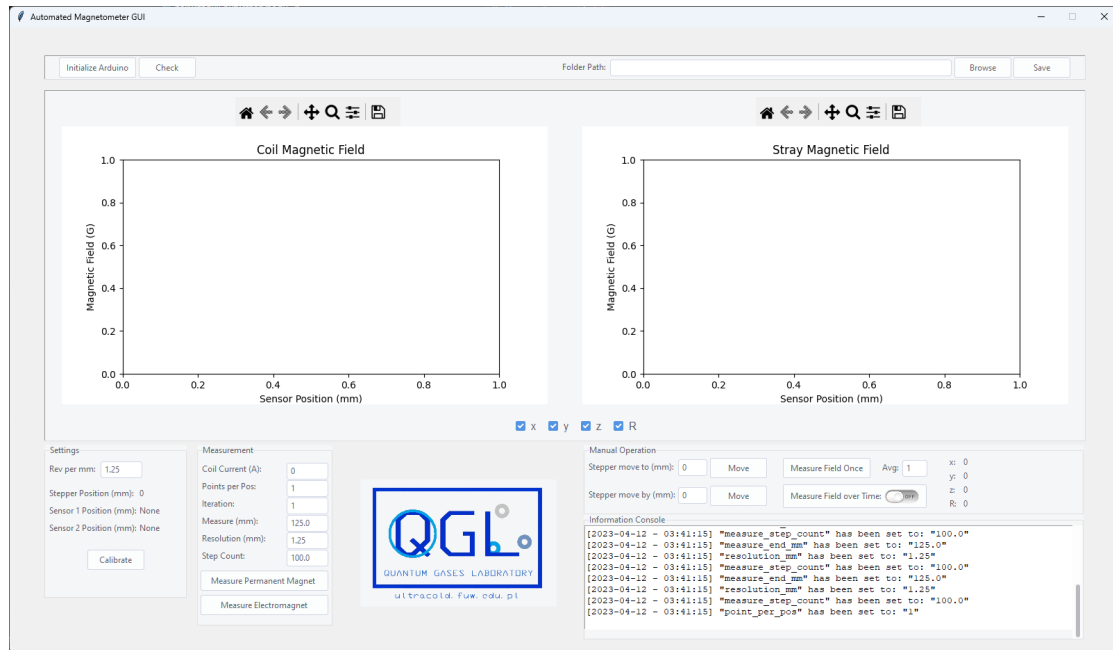


Figure 4.18: The graphical user interface of the automated magnetometer has designated sections for various functionalities. The top-left section is dedicated to establishing a connection with the microcontroller, while the top-right section is for specifying the data storage path. The two graphs in the GUI are employed for live plotting of measurement data: the left graph displays the magnetic field values of the electro/permanent magnet, and the right graph illustrates the stray magnetic field. The bottom part of the GUI is reserved for setting adjustments and manual operations.

It is also worth noting that the developed Python software facilitates advanced programming methods as the utilized firmware supports both synchronous (traditional) and asynchronous programming methodologies. Asynchronous operations, considered more advanced, allow tasks to run concurrently without blocking other tasks. This is achieved through techniques like event loops and callbacks, which enable tasks to be initiated and executed without waiting for their completion. Asynchronous programming enhances resource usage and responsiveness, particularly in scenarios involving external events or I/O operations.

On the other hand, synchronous programming executes tasks sequentially, leading to potential delays if tasks take a long time to complete, as subsequent tasks must wait their turn. Integrating sensors that operate solely with the I²C protocol into programs requiring fast readouts can be challenging. I²C communication involves a query-based protocol where the master device sends queries or commands to the slave device, which then responds with the requested data or action.

In synchronous programming, threading is often used to handle requested data, while asynchronous programming, with its efficient event loops, offers scalability advantages over synchronous threading. For tasks related to the Hall sensor and stepper motor driving, asynchronous

programming proves beneficial, especially for lengthy operations like I/O or network requests.

To compare the readout time of our Hall sensor with synchronous and asynchronous programming, we measured the time taken to retrieve readouts. After collecting 20,000 readouts in total, we found that synchronous programming takes an average of 186 ms per data point, whereas asynchronous programming takes 93 ms. These results highlight the significant reduction in data retrieval time achieved with asynchronous programming, making it ideal for real-time applications requiring fast readouts and offering flexibility in incorporating alternative hardware, such as different Hall sensor models.

The working principle of this magnetometer is based on the Hall sensor's polarization flip feature, where its magnetic sensing polarization direction is flipped. When a magnetic field source $\vec{B}_{\text{source}}$ is under examination, one needs to remember that the sensor's readout $\vec{B}_{\text{sensor}}$ is composed of three fields as:

$$\vec{B}_{\text{sensor}} = \vec{B}_{\text{source}} + \vec{B}_{\text{stray}} + \vec{B}_{\text{offset}}. \quad (4.12)$$

The $\vec{B}_{\text{stray}}$ in here is the present magnetic field exerted on the sensor apart from the $\vec{B}_{\text{source}}$. Thus, it includes other magnetic field sources and the stray field of Earth's magnetic field. The $\vec{B}_{\text{offset}}$ of the Hall Sensor is a variable that can fluctuate due to external magnetic fields or changes in temperature. We can utilize the polarization flipping feature to extract all needed fields with the following procedure:

1. without the magnetic field source in interest (noted as ns), measuring the sensor's response at each location y_i along the planned measurement path for two polarizations of the sensor,

$$\vec{B}_{\text{ns},+}^i = \vec{B}_{\text{stray}}^i + \vec{B}_{\text{offset}}^i, \quad (4.13a)$$

$$\vec{B}_{\text{ns},-}^i = -\vec{B}_{\text{stray}}^i + \vec{B}_{\text{offset}}^i, \quad (4.13b)$$

2. placing the magnetic field source (notes as ws) and repeating the measurement for the same locations y_i as before, for two polarizations of the sensor,

$$\vec{B}_{\text{ws},+}^i = \vec{B}_{\text{source}}^i + \vec{B}_{\text{stray}}^i + \vec{B}_{\text{offset}}^i \quad (4.14a)$$

$$\vec{B}_{\text{ws},-}^i = -(\vec{B}_{\text{source}}^i + \vec{B}_{\text{stray}}^i) + \vec{B}_{\text{offset}}^i. \quad (4.14b)$$

From the set of measurements described above, one can determine the field generated by the source, the stray field, and the offset field of the sensor for each measurement with the following:

$$\vec{B}_{\text{source}}^i = \frac{\vec{B}_{\text{ws},+}^i - \vec{B}_{\text{ws},-}^i - \vec{B}_{\text{ns},+}^i + \vec{B}_{\text{ns},-}^i}{2}, \quad (4.15a)$$

$$\vec{B}_{\text{stray}}^i = \frac{\vec{B}_{\text{ns},+}^i - \vec{B}_{\text{ns},-}^i}{2}, \quad (4.15b)$$

$$\vec{B}_{\text{offset}}^i = \frac{\vec{B}_{\text{ws},+}^i + \vec{B}_{\text{ws},-}^i}{2}, \quad (4.15c)$$

$$\vec{B}_{\text{offset}}^i = \frac{\vec{B}_{\text{ns},+}^i + \vec{B}_{\text{ns},-}^i}{2}. \quad (4.15d)$$

In some cases, the timings of these measurements might matter. For instance, a user might want to measure with and without the source in the same position before proceeding to the next one. Considering that, the magnetometer has two different measurement modes that can be called the *permanent mode* (for permanent magnets characterization) and the *coil mode* (for characterization of coils). In the *permanent mode*, firstly, the stray field in that region without magnets present is measured. Then, the measurement is repeated with the permanent magnets in place. In the *coil mode*, the coils do not need to be removed because a MOSFET-based power controller switch (ST1168 model¹) is utilized. This feature is implemented in the software, so the switching is automated. Here, the power switch turns off the current flowing through the coils for each measurement point to obtain the stray magnetic field (Equation 4.15b), then the switch turns on, and the magnetic field generated by the coils is measured (Equation 4.15a). The *permanent mode* can also be used to characterize coils if the user turns on the current flow once the stray fields have been measured. Under normal conditions, it is not expected to have significant differences between the two approaches, but they are distinctive if one looks carefully: in the *coil mode*, the determination of the stray field and the determination of the field from the coils in each location are temporarily separated by about 200 ms, whereas for the *permanent mode*, this could be even hundreds of seconds. This may play a role if stray fields are time-dependent, for instance, due to the activities of other people in the laboratory.

It is important to emphasize that even in the case where the characterization of the coils is made with a precise magnetometer, we encounter a unique challenge. The characterized coils will eventually be relocated to the main experimental setup, where any change in their characteristics will directly cause manipulation of the properties of ultracold atoms in an undesired way. In this new environment, achieving an accurate calibration presents a complex task, as having a moveable magnetometer inside the vacuum chamber will be absurdly complex. Therefore, an advanced in-situ calibration process is still vital, and in the best scenario, harnessing the ultracold atoms itself as our reliable testing ground.

One should note that the characteristics of the examined coils are expected to remain the same if such parameters as their dimensions (such as distance from one coil to another), temperature, or wirings are kept the same. However, in some circumstances, if the coils are attached separately to the experimental setup, it can still be subject to change. A good example can be seen in Table 4.2 where a comparison of the characterization with the Hall Sensor and the Zeeman spectroscopy is given. A recommended approach to such an issue is to have a rigid, non-magnetic mount to all sets of coils where their relative positions are preserved. Cabling is done in such a way that it does not differ from the measurements done in the initial characterization measurements. Nevertheless,

¹ST1168 is a microcontroller compatible power switch based on N-channel MOSFET IRF520N [151] by Infineon.

this might be impossible or not feasible, which is understandable due to the complexity of the ultracold apparatuses such as vacuum chambers or the need to have space around the coils for optical elements or laser beams themselves.

Another crucial gain with the initial characterization is to assess the coil's performance, eligibility, and validation of their defects, if any occurred during the manufacturing. The results shown in Figure 4.19 are great example of showing the off-axis (x and z) magnetic field is still present due to defects in the mounting or the coils itself, whereas expected there should be no field present.

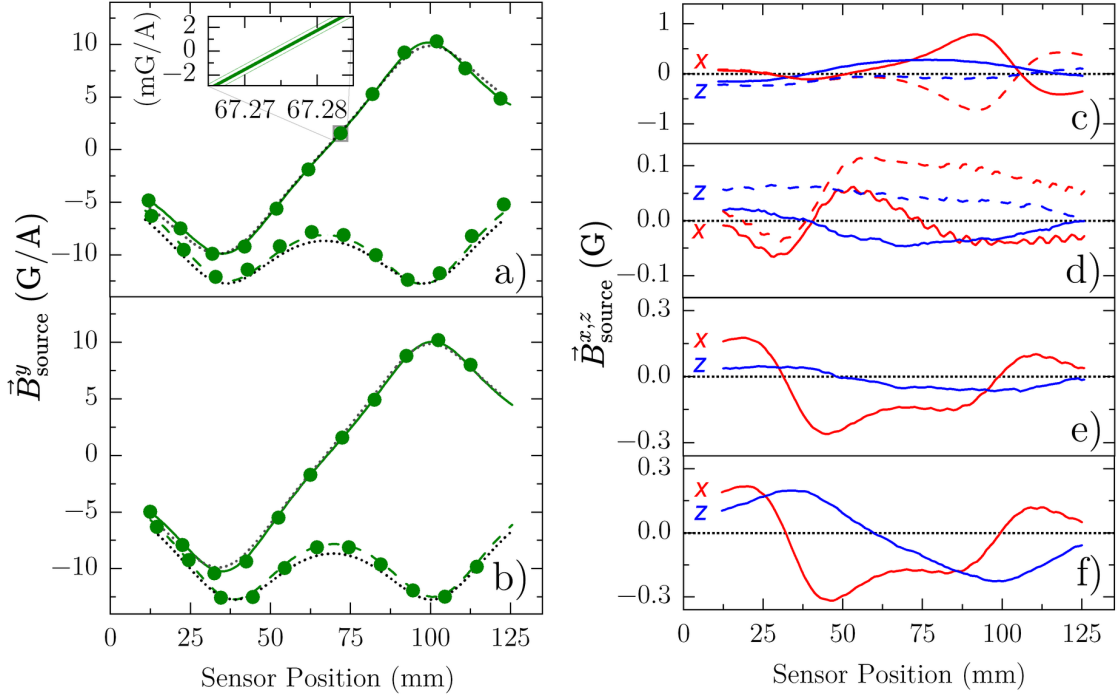


Figure 4.19: Magnetic field mapping of a set of coils shown operated in a Helmholtz configuration (dashed lines) and anti-Helmholtz configuration (continuous line) measured with the *permanent mode* with 30-point averaging. The data in a) and c) correspond to the coils assembled in the anti-Helmholtz configuration to operate as a source of a quadrupole field for a 2D magneto-optical trap; b) and d) coils individually aligned to minimize the deviations from the ideal anti-Helmholtz configuration; e) relative arrangement of coils as in d), with the assembly rotated by 1° around the z -axis; f) relative arrangement of coils as in d), with the assembly rotated by 1° around the z -axis and 1.5° around the x -axis. The inset in a) shows the confidence intervals set by the root-mean-square value of the measured data points. Magnetic field projections along the sensor axes x , y , and z are color-coded with red, green, and blue, respectively. The gravity points along the z -axis, and the y direction is along the translation displacement of the Hall probe. The green dots in a) and b) are measurements obtained with a commercial gaussmeter, as discussed in the main text. We compare our measurements with a simulation of the magnetic field created by the coils, shown with a black, dotted line. Here, the simulation does not take into account the misalignment of the coils with respect to each other or with respect to the measurement path.

Initially, as shown in Figure 4.19c, the coils produced a significant amount of off-axis magnetic field due to their suboptimal alignment with each other. Subsequently, the off-axis fields were minimized through precise adjustments of their angles relative to one another, as shown in Figure 4.19d. In the later stages of the experiment, intentional rotations of the coils around the z -axis and x -axis were conducted to monitor the variations in off-axis fields resulting from the misalignment of the magnetometer with the coil set. If significant defects within the set of coils are detected, it is best to address these issues before their installation on the experimental setup. Once the design and manufacturing of the coils are confirmed to meet the intended specifications, the next step involves the installation and subsequent post-installation characterization.

Advanced coil characterization (post-installation)

The advanced characterization phase involves employing more precise and rigorous testing methods following the installation of these magnetic field sources within the experimental apparatus. It relies on the known magnetic properties of ultracold atoms to precisely measure the magnetic fields generated by the installed electromagnetic coils. This characterization phase is dedicated to understanding how the coils function in the practical conditions of the experimental setup.

Two distinct approaches were employed during the advanced characterization phase. The first and most precise method was to measure the applied external field via RF transitions (Zeeman spectroscopy) in ^{41}K atoms. These experiments were conducted as part of the study led by Mateusz Bocheński [119].

The Zeeman spectroscopy method involves exciting transitions between Zeeman states using an RF wave. With the accurately measured magnetic dipole constants and hyperfine splittings for most alkali metals [45, 152, 153, 154], these transition frequencies between the Zeeman states are considered highly accurate in a given applied magnetic field. While in the absence of an external field, the Zeeman states are expected to remain degenerate, Zeeman states have magnetic dependencies on their energy levels. The analytical solution for the energy of a Zeeman state under an applied magnetic field amplitude is provided in the form of the Breit-Rabi formula [152, 155] and is presented in Equation 4.16 for ground state potassium atoms:

$$E(B) = -\frac{a^{hf}}{4} + g_I \mu_B m_f B \pm \frac{a^{hf} I(I+1/2)}{2} \left(1 + \frac{m_I x}{2I+1} + x^2 \right)^{1/2}, \quad (4.16)$$

where,

$$x = \frac{(g_J - g_I) \mu_B}{a^{hf} (I + 1/2)} B. \quad (4.17)$$

In this context, a^{hf} is the hyperfine splitting at zero applied magnetic field, g_S represents the electron g-factor, g_L stands for the gyromagnetic factor of the orbital (where $g_L = 1 - m_e/m_n$, with m_e denoting the electron mass and m_n the nuclear mass). Additionally, μ_B is the Bohr magneton, and the sign ($\pm$) corresponds to the manifolds with $F = I \pm S$. Furthermore, g_J is the Landé g-factor of the electron, and g_I represents the nuclear gyromagnetic factor.

The Zeeman spectroscopy of ^{41}K atoms was conducted in the following experimental sequence. The atoms were cooled down and transferred to our optical dipole trap (detailed previously in [Section 4.2.2](#)) with a temperature of approximately $\sim 18 \mu\text{K}$. Once loaded into the optical dipole trap, the atoms were pumped to the $F = 1$ state. Simultaneously, the current flowing through the atom dispenser was switched off to eliminate any potential additional stray magnetic field affecting the measurements. The RF wave was produced utilizing an RF signal generated by a Direct Digital Synthesizer (DDS), capable of producing frequencies ranging from 1 to 400 MHz. Subsequently, the signal was amplified using an RF amplifier (*ZHL-20W-202-S+*). The amplified signal was fed to an RF antenna configured to radiate within the 200-300 MHz range, directed toward the ultracold atoms.

Initially, to determine the existing stray magnetic field, all electromagnetic coils were turned off, and the frequency of the driving RF signal was swept across from 253.8 MHz to 254.2 MHz in 2 kHz increments, measuring allowed transitions between Zeeman levels from the $F = 1$ to $F = 2$ of the ground state. Subsequently, an RF wave pulse with these frequency steps was applied to the sample in an iterative sequence, each using the predetermined frequencies. Shortly afterward, the population of atoms in the $F = 2$ state was determined via absorption imaging. When the frequency of the driven RF wave matches the resonant frequency of a Zeeman transition between two states of an atom, the atoms are substantially pumped into the excited state ($F = 2$) from their initial state ($F = 1$). The results of this measurement are depicted in [Figure 4.20](#), revealing five distinct peaks corresponding to seven possible transitions between Zeeman levels of $F = 1$ ($M_F = -1, 0, 1$) and $F = 2$ ($M_F = -2, -1, 0, 1, 2$) states.

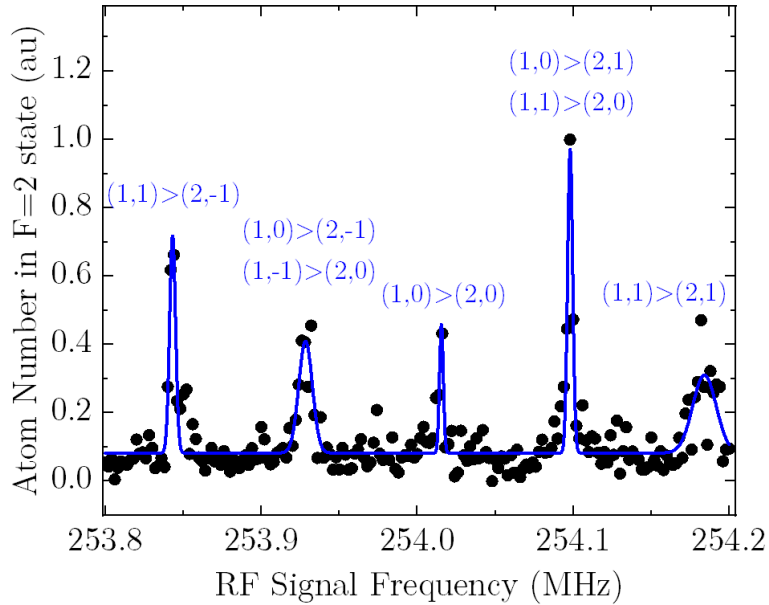


Figure 4.20: Zeeman spectroscopy was conducted on ^{41}K atoms within an optical dipole trap to measure stray magnetic fields. The measurement readout exhibits five peaks, and the corresponding transitions between Zeeman states are indicated with $(F, M_F) > (F, M_F)$ above the peaks. The blue line is derived from fitting a Gaussian function. The figure is based on data collected by M. Bocheński.

The calibration of the electromagnetic coils specifically utilized transitions $F = 1, M_F = 0 \rightarrow F = 2, M_F = 0$, and $F = 1, M_F = 1 \rightarrow F = 2, M_F = 1$, which are shown in Figure 4.20. The following procedure was applied to calibrate each set of coils. In each measurement, these particular transitions were selected, and the applied magnetic field via the selected electromagnetic coil was adjusted using a voltage-controlled current source. To prevent possible instabilities and enable independent determinations, every alternate set of coils was turned off, and only one was utilized at a time. When the desired magnetic field strength was reached, a pulse of RF wave generated via the installed antenna was passed through the sample. Similarly to the stray field measurements, the driving frequency was tuned iteratively to track the resulting population of atoms in the $F = 2$ state. By iterating this sequence for various control voltages (thus, applied magnetic field), the transition frequencies corresponding to the applied magnetic field strength were determined. The results were used in the fitting process according to Equation 4.16, and a relation between the applied control voltage (thus, current flowing through coils) versus the applied magnetic field amplitude was found.

Even though this first approach is superior to any other, as it relies on the narrowness ($\Delta\nu \leq 10$ kHz) of transitions between Zeeman levels in the ground state, it was implemented after the photoassociation spectroscopy measurements of the cesium atoms. Therefore, for the experiments described in this thesis (before the Zeeman spectroscopy), we have relied on an alternative calibration approach that utilizes the known location of magnetic Feshbach resonances of cesium dimer [50]. Later, when the Zeeman spectroscopy-based calibration was performed, it became possible to compare it with the method used throughout this thesis.

Furthermore, it may be queried why Zeeman spectroscopy was conducted using potassium atoms rather than cesium. This decision stemmed from the fact that the measurement exclusively utilized ^{41}K atoms, owing to its hyperfine energy splitting in the ground state levels, which is 254 MHz [152]. In contrast, for cesium atoms, this splitting amounts to 9.192631770 GHz (exact) [45] (refer to Figure 4.8), a frequency beyond the capabilities of the frequency generators accessible in the laboratory at the time.

In our alternative approach to calibrate our magnetic field sources, we focused on obtaining Feshbach resonance data for cesium atoms in particular atomic states, namely $(F, M_F) = (3, 3) + (3, 3)$ and $(3, 3) + (3, 2)$. A narrow segment of this data centered around 48 G is illustrated in Figure 4.21. This approach relies on the indirect measurement of atom collisional properties under specific conditions. It is essential for us to observe Feshbach resonances in our experimental setup, as they serve as crucial reference points for calibrating our electromagnetic coils.

The observability of the Feshbach resonances highly depended on the temperature of the prepared sample and the stability of the applied magnetic field [156]. Unfortunately, our attempts to observe Feshbach resonances in our setup initially yielded unsuccessful results. Our primary approach to observe the Feshbach resonances involved optimizing our setup to prepare colder samples with spin polarization, which is a prerequisite for further evaporative cooling and the eventual creation of a Bose-Einstein Condensate (BEC) [157]. In addition to this long-term approach, we sought to establish a relationship between the collisional properties of cesium atoms and the photoassociation process. Since the photoassociation process requires collisional wavefunction to overlap with a molecular state, tuning the collisional properties influences the rate of molecule formation [34]. This approach is often employed to enhance the probability of exciting colliding atoms into a molecular state; otherwise, the probability of this process (or the rate of molecule formation) is relatively low. In some instances, these enhancement methods

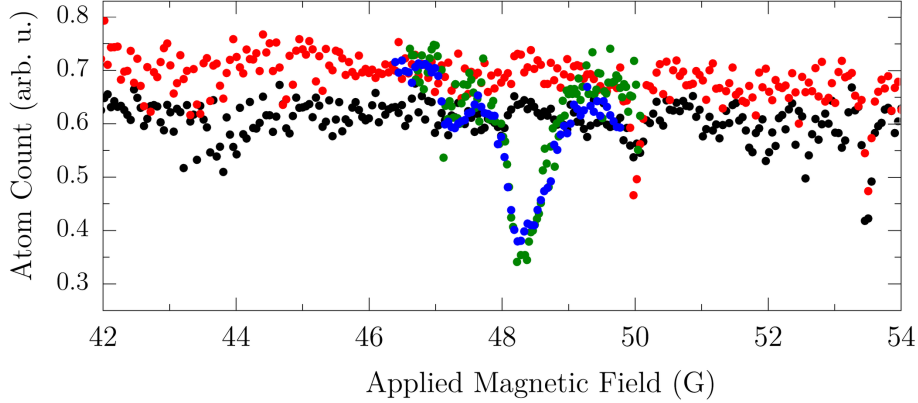


Figure 4.21: An example of a Feshbach spectrum obtained with blue detuned light and with light resonant to a photoassociation transition, which was used for magnetic field calibration. Feshbach resonances below 60 G using blue detuned light (blue) and via having a laser beam to be parked at a photoassociation transition to the vibrational level $\nu' = 233$ of the $1_g(P_{3/2})$ state (green) their roles in the measurement discussed later in this section. The ability to have spin-polarization of our sample can also be used to observe the Feshbach resonances of cesium atoms in the states $(F, M_F) = (3, 3) + (3, 3)$ shown with black data points and $(3, 3) + (3, 2)$ with red data points.

may even become necessary to observe weak photoassociation lines, as evidenced by the failed attempt in the work done in Ref. [23], but later successfully observed within this thesis work.

As a first step in the calibration process, we identified a strong photoassociation line corresponding to the vibrational level $\nu' = 233$ in the $1_g(P_{3/2})$ excited molecular potential of Cs_2 where $\nu' = [\nu_D - \nu]$. ν_D is the vibrational level numbering at the dissociation limit. This particular photoassociation line was relatively easy to observe in an optical dipole trap due to a very large photoassociation rate. For the rather moderate intensity of $\sim 14 \text{ W/cm}^2$, the photoassociation line was nearly 8 GHz broad, over three orders of magnitude broader than the natural linewidth typically associated with excited molecular states. A 1 second long photoassociation pulse would associate most atoms into molecules, consequently depleting the cesium atom population in the trap. For the sake of the measurement, we systematically reduced the power of the photoassociation beam until the atom loss within the dipole trap was no longer observed. Having a broad photoassociation line is crucial as it helps address concerns about the long-term stability of the tunable lasers. It also implies that there is no constant need to track the laser frequency, as it will only drift by a few tens of megahertz over the course of a few hours when stabilized with a reference cavity. Its broad feature also comes in handy when the Zeeman shifts are taken into account since magnetic fields up to several hundreds of Gauss will affect the photoassociation rate negligibly low. Undoubtedly, one can also choose a photoassociation line that is not broad, but then it becomes necessary to tune the laser frequency with each change of the applied magnetic field. In fact, if the frequency of the laser is known precisely (with optical referencing tools such as frequency comb), then one can track these changes to characterize the magnetic field coils by tracking the Zeeman shift similarly to the Zeeman spectroscopy method described above.

The characterization experiment follows the procedure outlined below. In each iteration, the sequence starts with loading atoms into the MOT using a conditional loading scheme. It involves

monitoring the fluorescence level of the atoms loaded into the MOT to trigger the next steps of the experimental sequence upon reaching a predetermined threshold, as described in [Section 4.5.1](#) with more details. Following the MOT stage, the CMOT (compressed-MOT) phase is executed to compress the atomic cloud, increasing atomic density and enhancing cooling efficiency, before proceeding to the GMC phase to further cool the atoms to 10 μK . Subsequently, the atoms are loaded into the dipole trap, where they can be spin-polarized in either the $F = 3, M_F = 3$ state or prepared as a mixture of $F = 3, M_F = 3$ and $F = 3, M_F = 2$ states, with a negligible number of atoms left in other spin-states (later detailed in [Section 5.3](#)). Now that the atoms are in the desired states, the magnetic field is ramped to a designated value. Then, atoms are exposed to the photoassociation light for typically 1 second. Finally, the magnetic field is ramped-down in a controlled manner to 0 G. Subsequently, a fluorescence measurement is performed to determine the remaining number of atoms in the dipole trap.

In these measurements, we have observed numerous Feshbach resonances that enhanced the photoassociation rate for the selected photoassociation transition. This enhancement reduced the remaining atom population in the optical dipole trap, indicating additional atom loss due to the increased molecule generation rate (green data points in [Figure 4.21](#)). We also employ a similar method to the photoassociation approach. In this method, we manipulate two-body radiative loss using an excitation laser that is blue-detuned from the cesium D_2 line [[31](#), [158](#), [159](#)]. By doing so, we couple the atom pair in the ground state to an electronically excited molecular state with a repulsive interaction. This leads to a relative increase in the kinetic energy of the atoms, ultimately ejecting them from the trap, leading to enhanced losses in the vicinity of Feshbach resonances (blue data points in [Figure 4.21](#)). These measurements were performed over a magnetic field range spanning from 3 G to 70 G, effectively identifying many of the Feshbach resonances for both $(F, M_F) = (3, 3) + (3, 3)$ and $(F, M_F) = (3, 3) + (3, 2)$ states. It is worth noting that the observed Feshbach resonances were achieved without relying on various other approaches, such as slow magnetic field ramping and rapid oscillations [[160](#)]. The parameters for these methods, including specific scanning speed or frequency of the rapid oscillations, often vary for each resonance. Our method eliminates the need to conduct such time-consuming and complex processes, streamlining the experimental procedure.

When measurements conducted in our apparatus enabled us to observe the Feshbach resonances of cesium, it presented an opportunity to roughly verify the initial characterization of the Feshbach coils. Also, it provided a chance to perform cross-calibration using compensation coils to calibrate the Feshbach coils (as introduced at the beginning of this section). Our compensation coils were already calibrated while developing a bidirectional, analog current source for our compensation coils [[161](#)]. In that work, the control electronics were benchmarked with stray field measurements by tracking the efficiency of sub-Doppler cooling of ^{39}K atoms in gray molasses. The cross-calibration process is utilized by varying the applied magnetic field from the already calibrated coils, and then compensating for this change by tuning the uncalibrated coils. Therefore, compensation coil values for each coil specified for the x , y , and z axes are extracted from measurements reported in Ref. [[161](#)]. [Table 4.2](#) presents the calibration results of a series of compensation coils designed to generate magnetic fields along the x , y , and z axes, in addition to the Feshbach coil, employing three distinct methods presented in this section.

Type	X Coil [G/V]	Y Coil [G/V]	Z Coil [G/V]	Feshbach Coil [G/V]
Hall Sensor	5.964(24)	8.051(31)	2.561(26)	106.26(112)
Zeeman spectroscopy	6.0294(51)	7.759(14)	2.5147(5)	102.142(59)
Cross-Calibration	6.09(3)	8.098(7)	2.51(9)	97.982(100)

Table 4.2: Table displaying the calibration results of a set of compensation coils generating magnetic fields on the x , y , and z axes, along with the Feshbach coil, using three different methods. The Hall Sensor method is done before installing the coils into the KCs single chamber. Zeeman Spectroscopy is done with measurements on ^{41}K , and cross-calibration is done using the data extracted on the experiments with ^{39}K .

The cross-calibration method was applied via the observation of Feshbach resonance at 47.97 G, and the position of the Feshbach resonance was tracked in the following systematic way. Firstly, a pre-selected compensation coil was ramped to an arbitrary value, and the Feshbach coil was ramped until the specified Feshbach resonance was observed. For a given magnetic field reduction with the offset coils, the Feshbach coils were ramped up iteratively with the minimum step allowed by the data acquisition (DAQ) card of the experimental control electronics. The peak point of the Feshbach resonance is then re-determined in units of applied current to both coils. This makes the additional magnetic field added by the Feshbach coil identical to the previously ramped down magnetic field amplitude by the already calibrated compensation coils:

$$|\vec{B}_{\text{FR}}| = |(\vec{B}_{\text{FR}} + \Delta\vec{B})_{\text{calibrated}} + \vec{B}_{\text{uncalibrated}}|, \quad (4.18)$$

$$|(\Delta\vec{B})_{\text{calibrated}}| = |\vec{B}_{\text{uncalibrated}}|,$$

where, B_{FR} represents the magnetic field where a Feshbach resonance is centered. The obtained data is fitted to determine the relationship between the control voltage and the corresponding magnetic field. (see Figure 4.22).

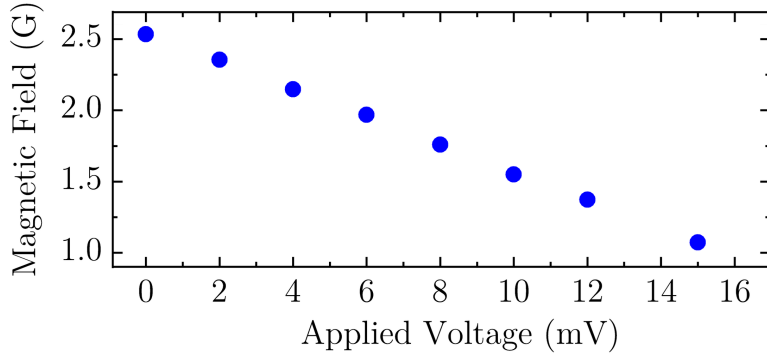


Figure 4.22: Cross-calibration method results were utilized to correlate the voltage supplied to the Feshbach coils with the magnetic field amplitude ramped down by the compensation coils. Initially, the compensation coils are gradually ramped down while being resonant with a known observed Feshbach resonance. Subsequently, the lacking magnetic field strength is applied in the same direction using Feshbach coils. The voltage difference is then determined employing the calibration values obtained for the compensation coils via Zeeman spectroscopy of ^{41}K atoms.

The cross-calibration being completed marks the conclusion of the advanced characterization phase to proceed with further experiments. One can now compare the acquired Feshbach Resonance data (below 70 G) to theoretical calculations (see Figure 4.23). It is important to consider that observed Feshbach resonances may shift regarding the applied magnetic field due to temperature variations. Thus, this benchmark analysis serves to guide users of this setup in estimating the deviation between the assigned field value and the actual field at which a Feshbach resonance appears, allowing for an estimated range of magnetic fields when studying a specific Feshbach resonance.

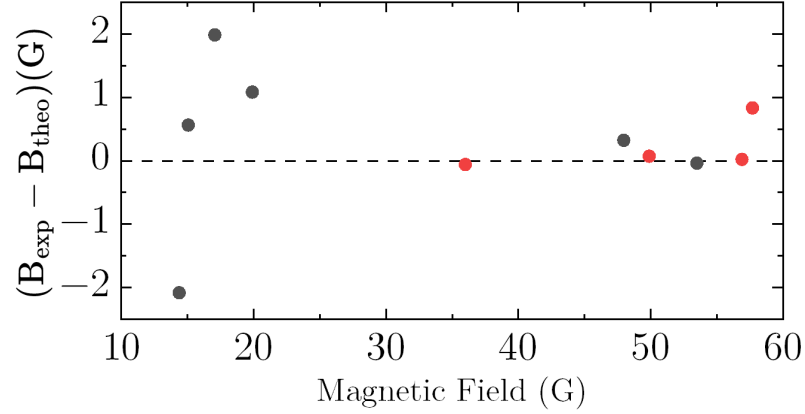


Figure 4.23: The deviation of measured Feshbach resonances (B_{exp}) from theoretical predictions (B_{theo}) [162]. Black data points represent measurements taken when the colliding atoms are in states $(3, 3) + (3, 3)$, and red data points represent measurements taken when the colliding atoms are in states $(3, 3) + (3, 2)$.

4.4 Imaging setup

The imaging setup used in our ultracold experiments is capable of performing both absorption and fluorescence imaging of ultracold cesium and potassium atoms. It is equipped with diffraction-limited optics in both the vertical and horizontal imaging paths for the given wavelengths used in the imaging of cesium ($\lambda_{\text{Cs}} = 852 \text{ nm}$) and potassium ($\lambda_{\text{K}} = 767 \text{ nm}$) atoms. The optical layout of the imaging setup is illustrated in Figure 4.24.

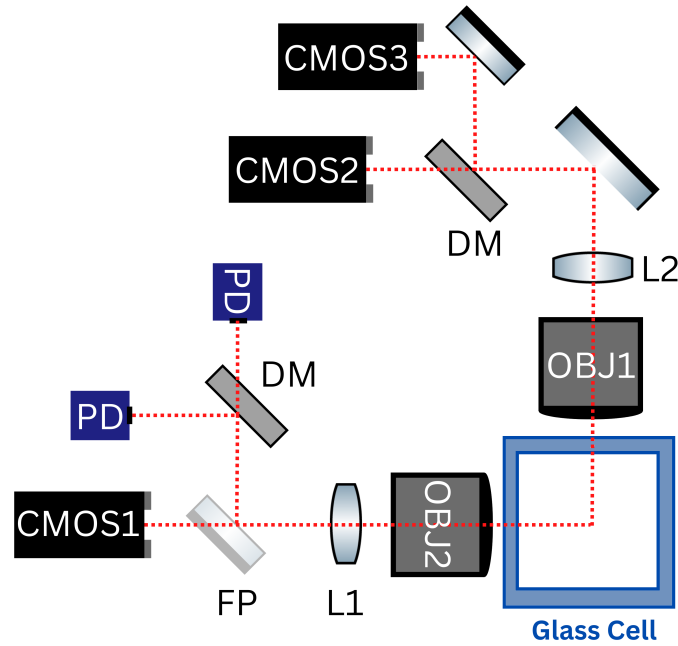


Figure 4.24: The horizontal (left) and vertical (top) imaging setups are designed for conducting absorption and fluorescence imaging of ultracold potassium and cesium atoms. Red dashed lines represent the optical axes for readout devices. OBJ1/2: Imaging objectives. FP: Mirror attached to a flip-able mount. PD: Photodiode. CMOS: Active-pixel sensor. DM: Dichroic mirror. L1/2: Achromatic lenses. *The optical components depicted in this figure are licensed under Ref. [121].*

The imaging setup has two paths dedicated to horizontal and vertical imaging. In the horizontal path, a CMOS camera facilitates the simultaneous imaging of both atomic species. In addition, two photodiodes are employed, enabling the simultaneous and distinctive detection of fluorescence signals emitted by cesium and potassium atoms. The distinctive detection is achieved with dichroic mirrors that split the light collected by the objectives into diverse paths distinguished by the species of the atom. All dichroic mirrors in the imaging setup exhibit high reflectance for light at a wavelength of 767 nm and high transmission at a wavelength of 852 nm. However, only one method can be employed at a time in this path, with the method selection determined by a mirror mounted on a flippable base. This mirror directs light into the photodiodes or, when moved out of the way, allows the light to be collected by the CMOS camera. This particular CMOS camera has been the primary source for most of the readouts in our ultracold experiments thus far. The imaging setup described herein facilitates the capture of ultracold atom readouts

for up to 20 ms subsequent to their release from the trapping region. This is achieved as the atoms persist within the field of view of the objective lens during their free fall. However, beyond this 20 ms duration, the atoms transition out of the field of view of the imaging objective. This capability is particularly a required feature of ultracold imaging setups, providing sufficient time to track the expansion of the ultracold sample and accurately determine its temperature, a typical measurement known as ‘time-of-flight’.

Similarly, we utilize two CMOS cameras and a dichroic mirror in the vertical imaging path, enabling simultaneous and distinctive imaging of both species.

The remainder of this section is dedicated to designing and characterizing our imaging objectives used in the setup. In contrast, absorption imaging is detailed in [Section 4.4.1](#), and the fluorescence detection scheme is provided in [Section 4.4.2](#).

The objectives, which are core elements responsible for collecting light from atomic samples, are constructed based on the designs introduced by Pritchard et al. [163]. In particular, our imaging setup features two distinct objectives (ATOM and TRAP in the Ref. [163]), referred to as OBJ1 and OBJ2, throughout this section. The designs reported by Pritchard et al. can be assembled using multiple commercially available lenses, providing the flexibility to adjust the spacing between the lenses to meet specific imaging requirements. This adaptability is particularly valuable in scenarios where conditions, such as the utilized wavelength, differ from those covered in the Ref. [163]. We opted to utilize these designed objectives because their working distances were compatible with our experimental constraints concerning the distance between the glass cell and objectives, often referred to as the working distance. Additionally, the objectives were designed using 2-inch lenses to meet our requirement of achieving the highest possible numerical aperture to enhance light collection emitted from our ultracold atoms.

These imaging objectives are infinity corrected and, in their default versions, have $NA = 0.175$ for OBJ1 and $NA = 0.29$ for OBJ2. OBJ1 provides a resolution of $3\text{ }\mu\text{m}$, while OBJ2 is optimized for high collection efficiency with a resolution of $1.8\text{ }\mu\text{m}$. These objectives are specifically tailored for the wavelengths of 852 nm and 802 nm, respectively. As mentioned earlier, they are constructed from multiple commercially available lenses. For the first objective, the lenses used include *Thorlabs* -LC1611, -LB1607, -LA1417, and -LE1418, while for the second objective include *Newport* -KPC067, *Thorlabs* -LB1199, -LA1256, and -LE1985 (depicted in [Figure 4.25](#)). The optical lenses composing the objectives have anti-reflection coatings that induce high transmission over the 650-1050 nm wavelength range.

Our assessment of the objectives proceeded in two steps. Initially, we conducted simulations to ascertain whether the objectives, designed for ultracold atom imaging by Pritchard et al. [163], required any adjustments for our setup. The original design assumed a specific glass thickness between the sample and the objective. Although our setup utilized the same type of glass (fused silica), there were slight variations in thickness. In the original design, the objectives are optimized to have glass thicknesses of 6.4 mm for OBJ1 and 3.3 mm for OBJ2, whereas, in our setup, the window thickness of the experimental chamber is 4 mm. Consequently, we considered modifications to ensure the maintenance of diffraction-limited performance and to verify essential properties such as numerical aperture and effective focal length.

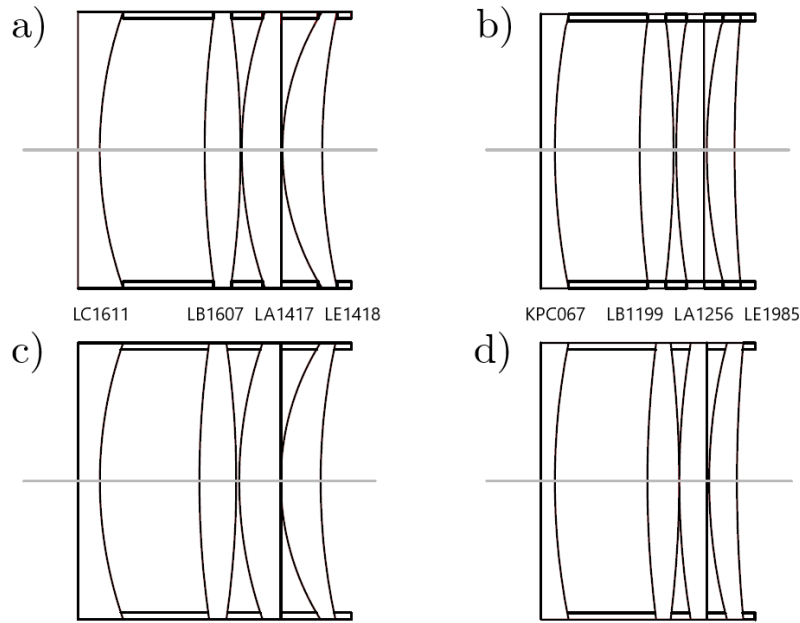


Figure 4.25: Cross-sectional view of the objectives: a) OBJ1, c) OBJ2, as reported by Ref. [163]. b) Modified version of OBJ1, and d) Modified version of OBJ2 designed for achieving diffraction-limited performance at wavelengths 767 nm and 852 nm with 4 mm of silica windows placed between the imaging objectives and the imaged plane.

When simulating an optical configuration, it is important to lay out the specific requirements and parameters for the intended task. Fortunately, a ray tracing simulation is often adequate for numerous optical design tasks, as optical systems typically exceed the scale of the wavelengths (λ) of the simulated light. In our case, the wavelength of the light emitted by the cesium and potassium atoms fulfills this criterion.

Typically, optical ray tracing software requires licenses for operation, and unfortunately, these licenses can be prohibitively expensive for individual users. In my experience, institutions often have limited licenses that can be shared among their staff. Regrettably, we have not had the opportunity to access licensed software such as Zemax, Code V, and FRED. However, there is currently a significant effort within the software engineering community to develop open-source alternatives, driven by the cost considerations mentioned earlier. This has drawn my attention to several Python libraries that provide optical design and simulation tools. While many are still in development and lack basic analysis tools such as wavefront error analysis or the ability to optimize optical setups with a given set of lenses, there is steady progress in this field. Recently, Stellar Software's optical design software, BeamFOUR [164], has made its content public, transitioning to open accessibility for users (a snapshot from the BeamFour software is shown in Figure 4.26). BeamFOUR is a ray-tracing-based simulation software designed for analyzing optical setups. It offers wavefront error analysis and optical design optimization capabilities by minimizing aberrations. Consequently, BeamFOUR is a valuable alternative to licensed software for similar purposes. It simplifies the determination of diffraction-limited optical systems and enables optimization of the setup with changes made to the configuration.

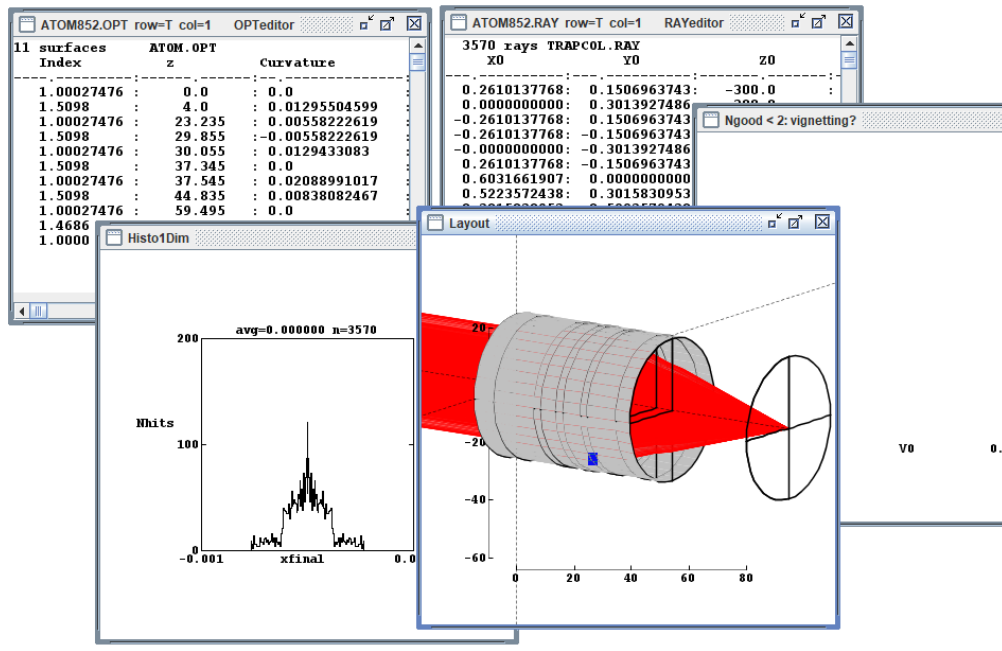


Figure 4.26: A snapshot of the BeamFOUR software [164] graphical user interface, wherein its built-in tools illustrate the second designed objective (OBJ2) under study. The display showcases rays originating from infinity and running parallel to the optical axis, as the objective is configured to be infinity-corrected. The GUI allows users to add optical surfaces with table-structured rows, and rays can be generated manually and via built-in functions provided by the software.

Even in the case of diffraction-limited optics, ray tracing is commonly employed to assess its geometrical aberrations. An optical system is deemed highly corrected if the RMS geometrical wavefront error is less than 7% of a wavelength [165, 166, 167], indicating that its performance is close to the optimal achievable for that aperture.

BeamFOUR's layout for optical components closely resembles that of other software, where each component is systematically listed based on its position along the optical axis. To validate the software's capabilities, I conducted an initial test by optimizing setups from a study by Ref. [163] on two distinct optical objectives, which they have presented in their work. Following successful iterations, I adjusted the thickness of the silica glass window to 4 mm, matching it with the thickness of our glass cell. Table 4.3 and Table 4.4 compare the original design parameters and the updated versions for our specific setup.

Surface	Curvature (mm)	Position (mm) Ref. [163]	Position (mm) This work	Material
1	∞	0.0	0.0	BK7
2	77.19	4.0	4.0	Air
3	179.14	23.235	22.45	BK7
4	-179.14	29.855	29.07	Air
5	77.26	30.055	29.97	BK7
6	∞	37.345	37.26	Air
7	47.87	37.545	37.26	BK7
8	119.32	44.835	44.55	Air
9	∞	59.495	59.25	Silica
10	∞	65.895	63.25	Vacuum
11	∞	115.095	114.33	Sample

Table 4.3: Design of the OBJ1 given with its surfaces of the optical elements and their positions on the optical axis. The table consists of the reference work’s lens placements and the optimized version with the change of the glass thickness.

Surface	Curvature (mm)	Position (mm) Ref. [163]	Position (mm) This work	Material
1	∞	0.0	0.0	BK7
2	129.2	2.5	2.5	Air
3	205.0	18.1	19.5	BK7
4	-205.0	24.3	25.35	Air
5	154.5	24.8	25.35	BK7
6	∞	29.9	30.45	Air
7	100.1	30.4	30.85	BK7
8	279.1	35.5	35.95	Air
9	∞	56.3	56.3	Silica
10	∞	59.6	60.3	Vacuum
11	∞	157.6	158.326	Sample

Table 4.4: Design of the OBJ2 given with its surfaces of the optical elements and their positions on the optical axis. The table consists of the reference work’s lens placements and the optimized version with the change of the glass thickness.

At the same time, in [Figure 4.27](#), the root-mean-square values of the wavefront errors are given as a percentage of the wavelengths 767 nm, 780 nm, and 852 nm. The wavelength of 767 nm is utilized for imaging ultracold potassium atoms, while 852 nm is for ultracold cesium atoms. The wavelength of 780 nm is provided in our analysis for comparison purposes, as discussed in the work of Pritchard et al. [163]. In this wavefront error analysis, a few different scenarios are inspected for both imaging objectives. Initially, OBJ1 was analyzed as in the work of Pritchard et al. [163] with the 6.4 mm of window thickness. Then, the window thickness was reduced to 4.0 mm, matching the thickness of our experiment chamber’s window thickness. Finally, the lens placements in this objective were modified to reduce the wavefront errors.

The distances between individual lenses were maintained with 3D-printed rings placed inside

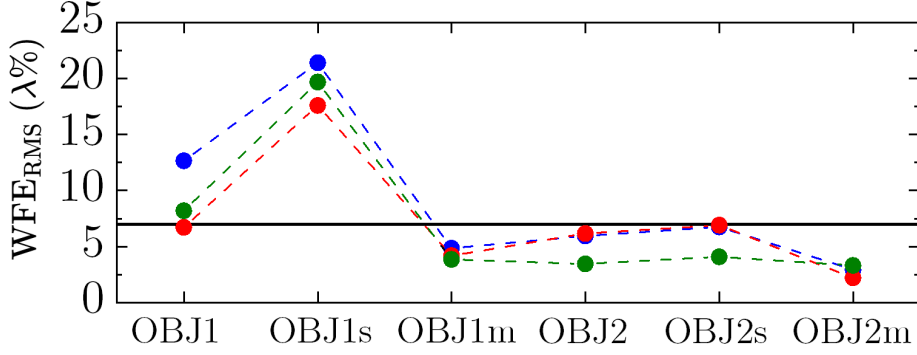


Figure 4.27: RMS of wavefront error (WFE) in percentage to wavelengths of 767 nm (green points), 780 nm (blue points), and 852 nm (red points) are given for different scenarios of the imaging setup. OBJ1/2: The imaging setup reported by Ref. [163] with a window thickness of 6.4 mm for OBJ1 and 3.3 mm for OBJ2. OBJ1/2s: The reference work’s objectives when simulated in our setup with the glass window thickness of 4.0 mm. OBJ1/2m: The modified and optimized versions of the imaging objectives when simulated in our setup. The black horizontal line indicates the 7% of the utilized wavelength.

a commercially available 2-inch lens tube to assemble the imaging objectives. The 3D-printed spacing rings were manufactured with a precision better than 0.05 mm, which is higher than the standards of conventional manufacturing precision. They were designed with smoothed edges to minimize the risk of damaging the lenses during assembly. To address potential dimensional errors that may arise during the 3D printing process, we employ abrasive paper to refine and ensure accurate dimensions of the prepared parts. It is recommended to use printing settings that prioritize precision over printing time, and for better, finer printing, consider using nozzles of 3D printers with smaller diameters.

After assembling the imaging objectives, we analyzed their performance using a setup that included a micro-ruler in the image plane, the designed objectives (OBJ1 and OBJ2 individually), and a secondary lens to project the image onto a CCD camera. The magnification of these infinity-corrected objectives depends on both the objective’s effective focal length and the secondary lens’s focal length employed for image formation:

$$M_{img} = \frac{f_{lens}}{EFL_{obj}}. \quad (4.19)$$

To determine the effective focal length, we introduced a sequence of secondary lenses with varying focal lengths of 250 mm, 500 mm, and 1200 mm. The lenses used were standard non-achromatic one-inch diameter lenses commonly employed in our laboratory; consequently, they were expected to exhibit higher wavefront aberrations than the achromatic lenses utilized in the installed version for imaging ultracold atoms.

The images captured with these lenses (an example is given in Figure 4.28) were utilized to identify local intensity maxima corresponding to the micro-ruler’s slits. Through this process, the average separation between each slit was determined in terms of pixels. With a known value of the pixel size of the CCD camera, we calculated the magnification of the optical setup.

Subsequently, by fitting the obtained parameters to Equation 4.19, we determined the effective focal length of the designed objectives to be 66.8 mm for OBJ1m and 120.2 mm for OBJ2m at 852 nm wavelength.

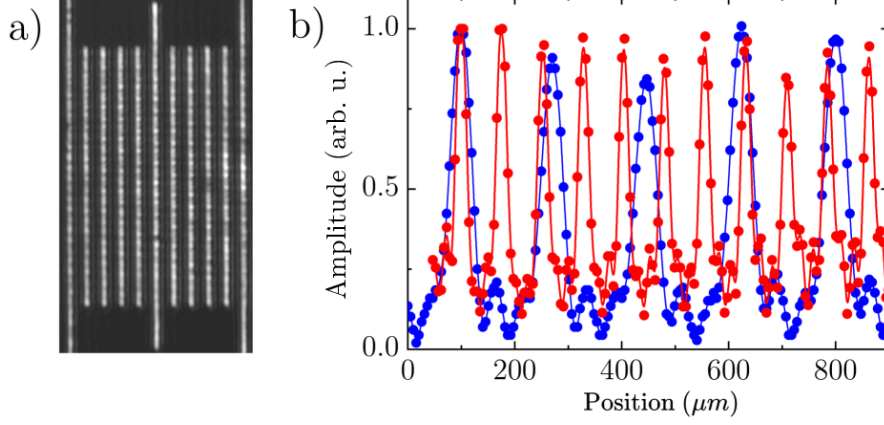


Figure 4.28: a) The micro-ruler is used to determine the effective focal lengths of the designed objectives. The image is taken with the OBJ1. b) A cross-section of a recorded image of a micro-ruler is displayed with light intensity versus position. Data points represent an image taken with the OBJ1 and a secondary lens with a 1200 mm focal length (blue) and 500 mm focal length (red).

Importantly, the effective focal lengths of the objectives were determined without silica glass positioned between the objective and the micro-ruler. Thus, given the 4 mm thickness of our experimental chamber, the effective focal length is expected to be larger (approximately +1.36 mm) than the determined values once the setup is installed. Nevertheless, the effective focal length is a valuable reference for estimating the desired focal length of the secondary lens. This estimation is crucial to align with the preferred magnification value of the imaging setup, later set at 150 mm for the horizontal imaging setup (where OBJ2 is installed) and 415 mm for the vertical imaging setup (where OBJ1 is installed), determined through the effective focal length measurements conducted in these experiments. Furthermore, the measurements with the micro-ruler enabled us to confirm the assembly of the objectives and identify any potential impurities or dust particles that might have gone unnoticed during the assembly phase.

Further calibration was performed after the installation of the setup to the KCs single chamber apparatus. This involved recording the free-falling ultracold cesium atoms and determining the central position of the cloud for a given time after the sample was released from the trap. In summary, the ultracold atoms were released from our MOT at time t_0 . In each iteration, we extend the fall time t_{fall} while measuring and determining the position of the cesium cloud. This iterative process allows us to calculate the distance traveled by the atoms during free fall using the equation $\Delta y(t_{fall}) = \frac{1}{2}gt_{fall}^2$, where $y(t_0) = 0$. By considering the imaging setup's magnification and expressing the cloud's central position in terms of pixel coordinates, the displacement $\Delta y(t_{fall})$ is initially determined in pixel coordinates. With the known pixel size of the CMOS camera, the resulting fall distance imaged on the sensor of the camera is then compared with the actual displacement of the atoms magnification is then determined ($M = \Delta y_{CMOS}/\Delta y$).

From these measurements, the magnification was found to be 1.27, with the given focal length of the secondary lens as 150 mm (Thorlabs - LA1417-B), and the effective focal length of the installed setup is determined to be 117.27 mm. A similar experiment was not done for potassium atoms but expected to give similar magnification values $M_{img} = 1.28$ at 767 nm wavelength.

The vertical imaging path is not used to determine the sample sizes and has not been calibrated so far. However, its magnification is expected to be around 6.21, allowing us to analyze the spatial profile of the prepared samples in greater detail.

In this chapter, I am solely responsible for conducting ray-tracing simulations, implementing modifications to the imaging objectives, and overseeing the assembly and integration of the imaging setups. However, collaborative efforts from all members of our research group contribute to establishing subsequent sections detailing absorption imaging and fluorescence imaging setups. Notably, I independently lead the installation of the horizontal imaging path and the entire lock-in detection scheme. Nonetheless, it is crucial to recognize that other team members are involved in the calibration and some subsequent utilization of these setups post-integration, as emphasized throughout the text.

4.4.1 Absorption imaging

The readouts of the ultracold experiments provide crucial insights into the prepared atomic and molecular samples by determining their total counts and varying temperatures. These parameters serve as key indicators, influencing the determination of various ultracold phenomena. They play a pivotal role in understanding the relations between particles within an ensemble and their interaction with light, as these relationships are deciphered through the analysis of these parameters. For instance, collisional properties are determined by comparing the initial and final counts of the prepared samples under varying conditions. Additionally, the efficiency of any cooling method is assessed by determining the density of the sample, a parameter directly related to the temperature. Another advantage of absorption imaging is its ability to probe the internal state of the atoms, such as their spin or energy level. Utilizing lasers targeting different atomic transitions enables the selective excitation of various states of the atoms. This approach allows for determining the population of the energy states under study.

In contrast, the absorption imaging method offers numerous advantages, including its rapidness and the option for energy-state-selective measurements. This method is by far the most utilized technique in ultracold physics. It relies on the absorption of resonant light by the atomic ensembles and subsequent evaluation of their optical density by the spatial readouts of the transmitted light.

In a typical procedure, atoms are commonly illuminated with collimated resonant light with a beam size larger than the atomic ensemble. The plane where the atoms are present is then imaged, and the resulting intensity of the transmitted light is recorded. The absorption of light by atoms results in observing a shadow in the recorded image. The optical density is then determined by evaluating the initial intensity of the light (or, preferably, the intensity obtained in the absence of atoms along the light path) and the intensity of the light after passing through the sample. The transmission of the light through this sample can be as expressed by Beer–Lambert law:

$$I = I_0 e^{-OD}, \quad (4.20)$$

where, I is the transmitted intensity, I_0 is the initial intensity and OD is the optical density. For ultracold atoms, the optical density can be expressed as the atom column density $n(x, y)$ and the absorption cross-section σ where $OD = n(x, y)\sigma$. The absorption cross-section:

$$\sigma = \frac{\sigma_0}{1 + 4\left(\frac{\Delta}{\Gamma}\right)^2 + \frac{I_0}{I_{sat}}} \quad (4.21)$$

is determined by the atomic parameters; saturation intensity I_{sat} , the natural decay rate of the excited state $\Gamma_{D_2} = 2\pi \cdot 5.234(13)$ MHz and the detuning of the laser from the atomic resonance Δ [45], whereas the σ_0 the on resonance absorption cross-section is $\sigma_0 = \hbar\omega\Gamma/(2I_{sat})$. The parameters I_{sat} and σ_0 vary depending on the polarization of the resonant laser beam and the state to which the atoms are excited, in which their values are reported in the Ref. [45]. In practice, the above equations can be efficiently used only if the resonant imaged beam is not overexposed and the sample does not fully absorb the incoming light along the optical path.

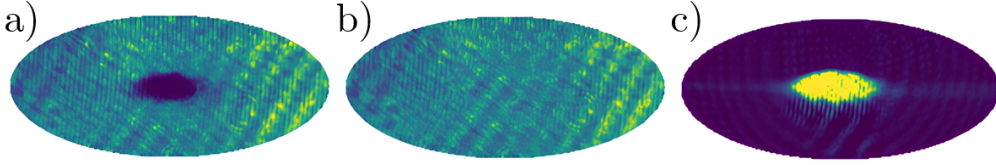


Figure 4.29: A series of images captured using the absorption imaging technique with the horizontal imaging setup. a) The initial image captures cesium atoms released from the crossed dipole trap, creating a shadow due to light absorbed by the atoms. b) An image of the imaging light without the presence of atoms. c) The subtraction of the first two images representing the imaged cesium atoms. All images in this sequence are normalized for enhanced visibility.

In contrast, the imaging sequence should obtain two readouts, as stated earlier, one with the presence of atoms I_a and one without their presence I_0 . Here, we already assume that the obtained images are subtracted from the obtained background images ($I_{a/0} = I'_{a/0} - I_{bg}$). A series of images is given in Figure 4.29 taken during an absorption imaging of cesium atoms in an optical dipole trap. Once these readouts are obtained, the optical density can be evaluated via the expression:

$$OD = \ln\left(\frac{I_0}{I_a}\right). \quad (4.22)$$

To determine the total atom count, one can integrate the optical density over the obtained data resolved across the image:

$$N_{atom} = \frac{1}{\sigma} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} OD(x, y) dx dy. \quad (4.23)$$

Our experiments typically employ a designated area on the recorded images, known as the region of interest (ROI). Atom counts are determined exclusively within this area, eliminating the need

for extensive computation across the entire image. This approach minimizes the impact of background noise outside the region where atoms may be present, preventing potential interference with the determination of atom counts. Note that this background noise can only increase the determined atom count, as the given equations do not anticipate any amplification of incoming light by the presence of atoms along the path. Therefore, the detected excessive light can be considered as noise in this case. However, this assumption cannot be made when the noise is present in regions where the light is already absorbed by atoms.

Determining the temperature of the ultracold samples requires several iterations of sample preparation under identical experimental conditions. To determine the temperature, the initial cloud size w_0 is first determined by an absorption imaging readout of the sample at the moment in which the cloud is released from the cooling trap. Subsequently, a series of iterations with an identical experimental sequence is conducted, varying only the time between releasing the atoms from the cooling trap and the absorption imaging. This allows for the determination of the expansion of the cloud over time. The variation in the cloud size over time is then expressed in terms of temperature using the Boltzmann distribution, given as [168],

$$w(t) = \sqrt{\frac{k_B T}{m} t^2 + w_0^2}, \quad (4.24)$$

where, k_B is the Boltzmann constant, m is the mass of the atom, and T is the temperature. The profile of the atomic ensemble is assumed to be Gaussian.

The atom count determination, and the temperature determinations are done via a post-experimental data processing platform made by P. Arciszewski and are available in Ref. [169].

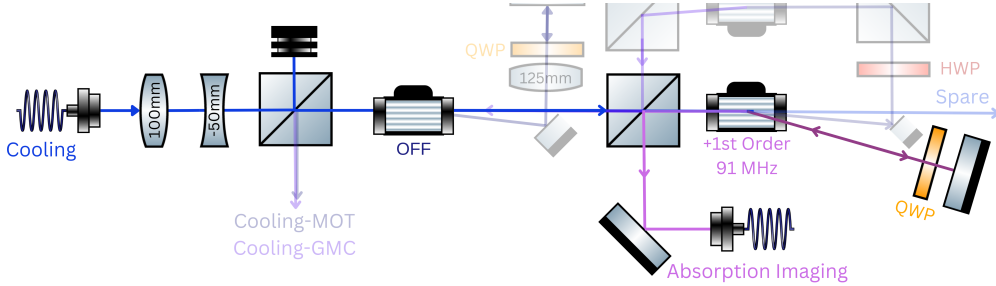


Figure 4.30: Layout of the setup used for preparing the absorption imaging light. The output from the cooling laser is guided to an AOM with a double-pass configuration. The prepared light is sent to the experimental chamber via polarization maintaining optical fiber. *The optical components depicted in this figure are licensed under Ref. [121].*

Now that the fundamentals of absorption imaging have been outlined, the remaining parts of this section will focus on our installed imaging setup, starting with the generation of the absorption imaging beam. The purpose of this setup is to prepare the absorption imaging beam in a way that ensures it has the desired frequency (frequency of the transition $6s^2S_{1/2}(F=4) \rightarrow 6p^2P_{3/2}(F=5)$) and power during the experimental sequences. The initial preparation of the light preparation setup for cesium was detailed in Section 4.2.1, where the Repumper and Cooling

lasers were first mentioned. The beam of the cooling laser in this preparation setup is directly guided through an AOM with a double-pass configuration driven at 91.5 MHz (see Figure 4.30). This driving frequency is generated by a DDS, and its output is amplified by an RF amplifier. The amplification ratio is set to the desired power values depending on the particular AOM's specifications.

The output from the double-pass AOM is then coupled into an optical fiber delivered to the optical breadboard located near the glass cell (see Figure 4.31). There, the absorption imaging beams for potassium and cesium atoms are overlapped on a PBS, with half-waveplates used to control the maximum amount of power that is sent for imaging. The absorption imaging light is then directed through the glass cell, either via a horizontal or vertical imaging path. Unfortunately, since this measurement is destructive, only one axis at a time can be used. A half-waveplate and a PBS are used to choose the desired path, and the angle of the waveplate is tuned to direct the light into one of these paths while the unused one is blocked physically.

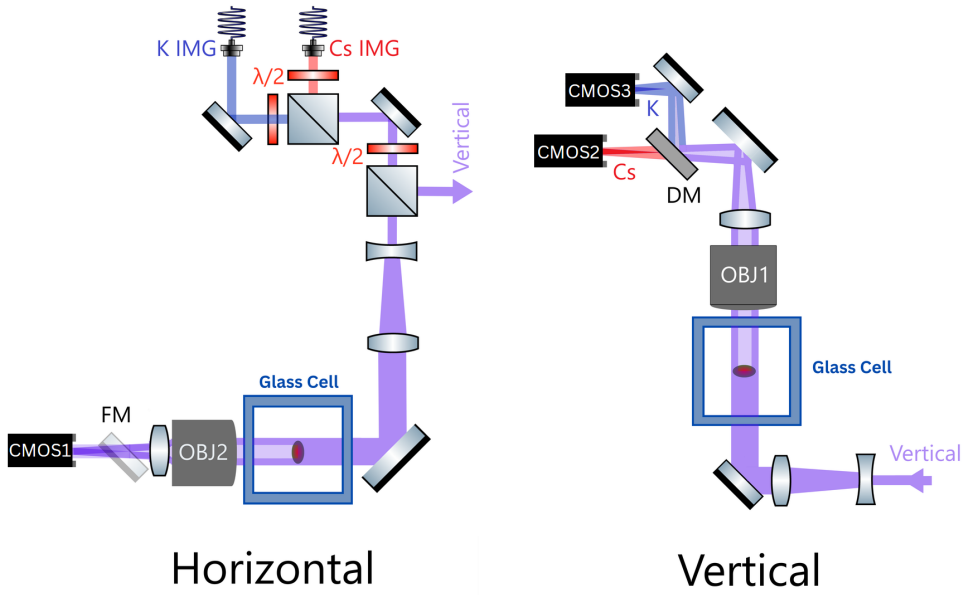


Figure 4.31: Horizontal and vertical imaging setups installed to our KCs single chamber apparatus. In the diagram, DM refers to the dichroic mirror, FM represents the flippable mirror, and OBJ1/2 corresponds to the imaging objectives. The readout devices are labeled as CMOS1/2/3, representing charge-coupled devices. *The optical components depicted in this figure are licensed under Ref. [121].*

In the horizontal imaging path, we utilize 'OBJ2,' introduced in Section 4.4, designed to achieve diffraction-limited performance. Following this infinity-corrected imaging objective, a lens with a focal length of 150 mm (LA1417-B) is employed to form an image of our ultracold samples on our CMOS² camera sensor. In our cesium experiments, the absorption imaging setup is designed to measure atoms in the $F = 4$ ground state energy level since it is done via excitation to $F' = 4$ state followed by the decay to $F = 4$ state. However, in most cesium experiments, atoms are

²Grashopper GS3-U3-41C6NIR-C by FLIR

initially pumped to the lowest energy level, $F = 3$, of the ground state. Consequently, before the absorption imaging process, atoms are repumped to the $F = 4$ state for imaging (refer to Figure 4.32) by illuminating the MOT repumper beams for a short 0.2 ms exposure time. Subsequently, a pulse of the absorption imaging beam is delivered, typically for 50 μ s, along with a trigger signal sent to the CMOS camera for image acquisition.

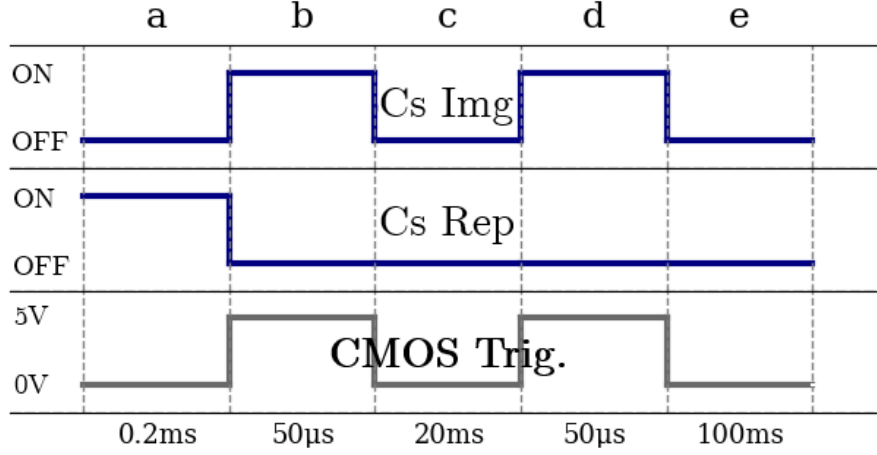


Figure 4.32: Sequence of absorption imaging used on ultracold ^{133}Cs atoms. a) Cs atoms in the $6S_{1/2}^2$ ($F = 3$) state are pumped into the state $6S_{1/2}^2$ ($F = 4$) by using the Cs repumper beam. b) A pulse of absorption imaging light is illuminated on the sample, and a trigger signal is sent to the CMOS to record the image. c) A waiting period for the cloud to leave the field of view by free-fall together with the time required for another image to be taken via the CMOS camera d) Second image for the absorption imaging is taken again with a pulse of absorption imaging light. The light is unaffected by the sample because the cloud is outside of the imaging system's field of view. e) Sequence is held for 100 ms to allow devices to return to their 'idle' states, whereas the recorded images are sent to the post-analysis software to be stored and processed.

After the imaging beam pulse, a waiting period of 20 ms is included in the sequence. This 20 ms interval is the minimum time required to capture two consecutive images using our CMOS camera. Importantly, it provides enough time for free-falling atoms to exit the field of view of the horizontal imaging setup. Following this 20 ms hold time, a second image is recorded by delivering another pulse of absorption imaging light through the sample for 50 μ s. The sequence is then held for the next 100 ms, during which the recorded images are sent to the software responsible for post-analysis [169], and all devices are set to their 'idle' state.

Before each imaging sequence, a trigger signal is sent to the CMOS cameras to clear any undesired charge accumulated on its pixels. This initial image capture occurs around 20 ms before the trigger signal is sent to carry the first absorption imaging readout. The timing is kept to a minimum, signifying the time necessary to record the next image. Thus, it minimizes potential pixel charging, primarily attributed to ambient temperature fluctuations and light exposure on the camera chip during idle mode.

4.4.2 Fluorescence imaging

Fluorescence imaging, another commonly used technique for imaging ultracold atoms, relies on collecting photons emitted by atoms continuously excited with resonant light. Ideally, the excited transition should be closed, meaning that the electron, after excitation, would return to the initial state. In alkali atoms, it is necessary to use two frequencies to achieve this, a feature discussed in previous sections in the context of magneto-optical trapping (Section 4.2.1). We utilize these two light frequencies, namely cooling-MOT and repumper-MOT beams in which the cooling-MOT beam is red-detuned 12 MHz from the $F = 4 \rightarrow F' = 5$ transition and repumper-MOT is red-detuned 1.24 MHz from the $F = 3 \rightarrow F' = 4$ transition for the fluorescence measurements. The ensuing fluorescence signal is subsequently either imaged using CMOS cameras or detected through photodiodes, utilizing the same imaging setup described in the absorption imaging section (Section 4.4.1).

In the experiments detailed in this thesis, fluorescence imaging serves as the primary method for assessing the relative change in atom count. Consequently, the emphasis is on relative atom numbers rather than absolute values. In cases where determining the absolute number of atoms is necessary, absorption imaging was utilized. The absolute atom count can, indeed, be determined by using fluorescence imaging too. Fluorescence imaging stands out as a primary method for quantifying atom numbers in small MOTs [170], quantum gas microscopes [171], and optical tweezers [172]. Its versatility extends to laser-cooled ions, where atom counts can only be determined through fluorescence readouts [173, 174]. When the system is thoroughly characterized, fluorescence imaging often surpasses absorption imaging in aspects related to atom number determination.

The determination of the atom number can be accomplished in the following way: the photons emitted during the fluorescence process are directly correlated with the atom scattering rate, which represents the rate at which an atom absorbs and re-emits incident photons. In our MOT, we utilize six cooling and six repumper beams. Here, we assume that only cooling beams exclusively contribute to the scattering process, as discussed in Ref. [45]. Consequently, the scattering arising from the repumper transition ($F = 3 \rightarrow F' = 4$) is not taken into account in this analysis. Under this assumption, the scattering rate, derived from a two-level system, can be expressed as:

$$R_{\text{sc}} = \left(\frac{\Gamma}{2} \right) \frac{(6I_0/I_{\text{sat}})}{1 + 4(\Delta/\Gamma)^2 + (6I_0/I_{\text{sat}})}. \quad (4.25)$$

Here, Γ represents the natural linewidth of the excited state with a specific value for cesium ($\Gamma_{D_2} = 2\pi \cdot 5.234(13)$ MHz [45]). The lasers operate at a detuning Δ of -12 MHz from the atomic resonance. I_0 signifies the intensity of a single cooling beam, and the isotropic saturation intensity I_{sat} is determined as 2.7119(69) mW/cm² [45]. The scattering rate can then be used for the evaluation of the atom count via the properties of the imaging setup. The expression for the atom count estimation is

$$N_{\text{atom}} = \frac{1}{R_{\text{sc}}} \frac{N_{\text{counts}}}{t_{\text{exp}} \eta_{\text{count}}} \frac{4\pi}{d\Omega}. \quad (4.26)$$

In this context, t_{exp} denotes the exposure time of the CMOS camera, and it can be adjusted between $3 \mu\text{s}$ and 7.03 s with our CMOS cameras [175]. η_{count} represents the camera's quantum efficiency, measured in counts per photon. The term $4\pi/d\Omega$ signifies the ratio of captured light, considering that fluorescence is emitted in all directions, yet only a fraction of the emitted light is collected. Here, $d\Omega$ corresponds to the solid angle, defining the extent of the field of view from a specific point that a given object encompasses. The solid angle, measured in steradians, ranges from 0 and 4π . With our horizontal imaging setup (illustrated in Figure 4.33), we can only collect approximately 3% of the fluorescent light emitted from the sample, whereas with our vertical imaging setup, we can collect around 5%. The quantum efficiency of our utilized CMOS cameras is 31.6% at 852 nm (cesium) and 45% at 767 nm (potassium) [175]. The atom count estimation (Equation 4.26) usually yields a reasonable number within an order of magnitude [176].

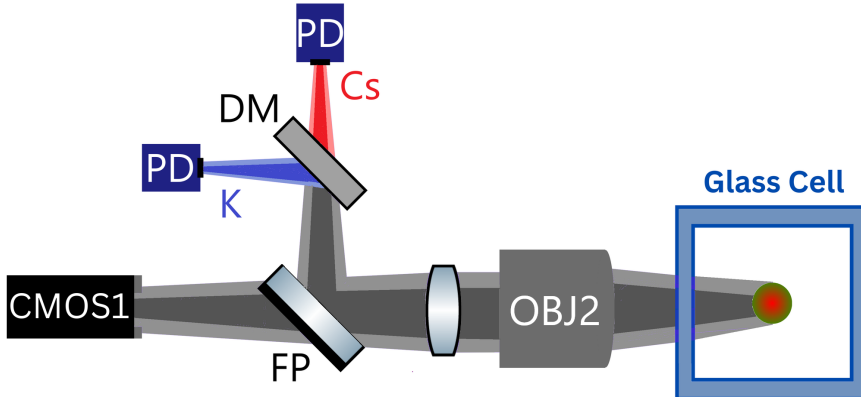


Figure 4.33: Illustration of the horizontal imaging setup for fluorescence imaging. OBJ: Designed objective for horizontal imaging. FP: Mirror attached to a flip-able mount. PD: Photodiodes. CMOS: Active-pixel sensor, DM: Dichroic mirror. *The optical components depicted in this figure are licensed under Ref. [121].*

Fluorescence imaging can also be used in measuring temperatures, particularly when employing techniques such as dropping the cloud through a light sheet [177, 178, 179] and analyzing either with fluorescence (or absorption) imaging. The temporal shape of the signal facilitates accurate temperature determination. However, challenges arise in very dense samples due to the potential re-absorption of photons, which limits the imaging, especially when obtaining the spatial distribution, which is crucial.

When evaluating our MOT, we can precisely determine the spatial distribution of the sample. However, to determine the temperature of the sample, time-of-flight measurements are necessary. This involves releasing the sample from the trap, thereby eliminating position-dependent and velocity-dependent forces acting on atoms. This is achieved by turning off the electromagnetic coils responsible for generating the magnetic field gradient (quadrupole field) to eliminate position-dependent forces and turning off the MOT beams to eliminate velocity-dependent forces.

Upon doing so, the atoms are released from the trap, and their temperatures can be determined by capturing a series of images with varying time allowed for the atoms to expand (time-of-flight), as explained in Section 4.4.1. Unlike absorption imaging, where a frame is taken with

an exposure time of $50\ \mu\text{s}$, our fluorescence imaging generally requires more than 2 ms of exposure time to collect a reasonable number of photons from our dilute samples. Consequently, during this exposure time, the cloud continues to expand while being influenced by the MOT beams illuminating the sample, leading to heating as atoms experience recoil upon absorbing or scattering photons. Hence, the recorded frame will likely display a different spatial profile than the actual one. This discrepancy arises because the frame incorporates photons emitted when the cloud was in both its initial and final sizes within the exposure time, along with the effects of sample heating. This potential discrepancy could introduce measurement errors, leading us to exclusively utilize absorption imaging to determine the prepared samples' temperature. This issue is often addressed by utilizing light sheets – a laser beam with a thin cross-section, where the atoms are profiled often in 2D with imaging setups suitable for low signal readouts even individual atom readouts were established on Rb atoms [177]. Our vacuum system does have mirrors inside to allow for such measurements, but it has not been utilized thus far.

The advantage of fluorescence imaging lies in the consistency of the readouts when using longer exposure times of the CMOS cameras. Although absorption imaging is used for atom number determination, fluorescence measurements offer a much higher signal-to-noise ratio. Absorption imaging readouts often exhibit interference effects on images due to reflections between the chip's pixels and the protection window in front of the CMOS sensor (refer to Figure 4.29 for visible interference patterns). In an ideal environment, this should not be an issue since interference patterns cancel out when subtracting images taken with and without the presence of ultracold atoms. However, the experimental setup includes shutters causing vibrations, coils generating high magnetic fields leading to coil mounts vibrating, and cooling fans of some devices causing micro-vibrations in the experimental setup. While these effects can be minimized with better device alignment or timing, they can never be eliminated.

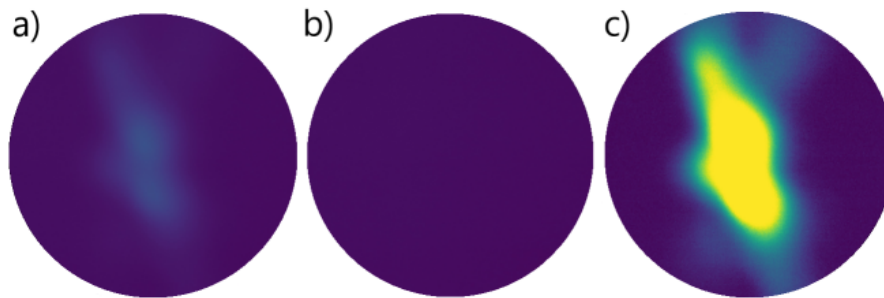


Figure 4.34: A series of images captured using the fluorescence imaging technique with the vertical imaging setup. a) The initial image captures cesium atoms released from the optical lattice. b) An image taken without the presence of the sample. c) The subtraction of the first two images resulted as an image of the cesium atoms. All images in this sequence are normalized for enhanced visibility.

The experimental sequence for fluorescence imaging, apart from imaging the operational MOT, is illustrated in Figure 4.35, closely resembling the absorption imaging sequence (as shown in Figure 4.32). The sequence begins with utilizing shutters to unblock the MOT beams, followed by a brief waiting period of approximately 0.1 ms to ensure that the shutters are fully opened. These shutters are essential for our experiments as they block and prevent leakage light from AOMs from reaching the ultracold sample.

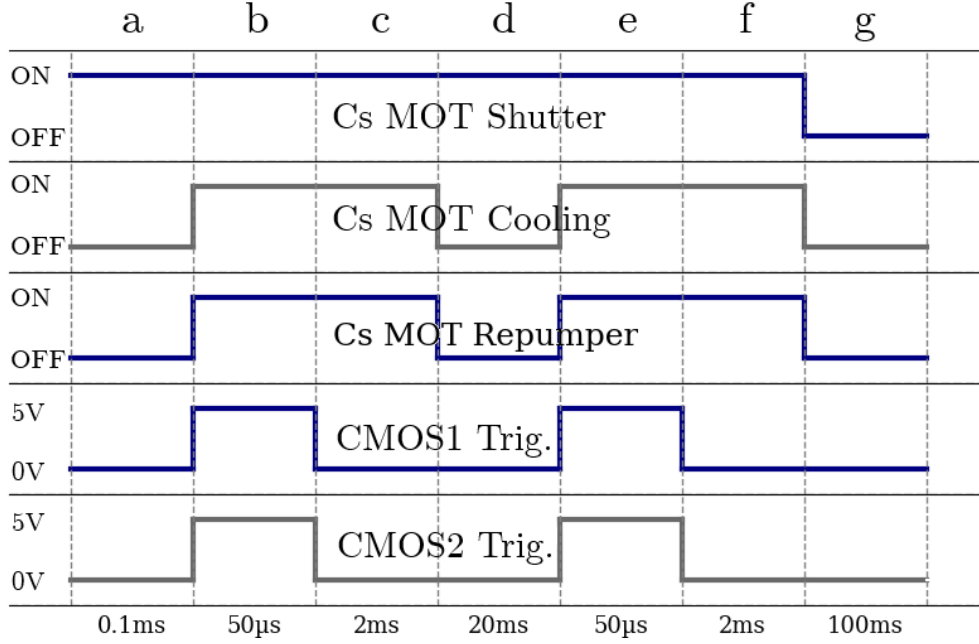


Figure 4.35: A typical experimental sequence for fluorescence imaging. a) The MOT shutters for cesium cooling and repumper beams are opened. b) Trigger signals to CMOSs are sent, c) together with the start of the illumination of the cesium cooling and repumper beams. d) 20 ms of holding time required for the next frame to be obtained via the same CMOS camera e) & f) A second image is taken this time without the sample presence due to the sample being out of the field of view of the imaging setups. g) Sequence is held for 100 ms to allow devices to return to their 'idle' states. If the MOT is not the subject of the imaging, the magnetic field gradient is not employed.

Subsequently, the cesium cooling and repumping beams are let to illuminate the sample for typically 2 ms, accompanied by digital voltage signals sent to the CMOS cameras for the start of the recording and data acquisition. In fluorescence imaging, both CMOS cameras (horizontal and vertical) can be simultaneously utilized since fluorescence signals are emitted in all directions. After capturing the first image and a waiting period of 20 ms, a background image is acquired. The duration of 20 ms corresponds to the necessary time required for our CMOS cameras to record subsequent images. Finally, the sequence is held for the next 100 ms in which the recorded images (as in Figure 4.34) are sent to the software responsible for post-analysis [169], and all devices are set to their 'idle' state.

'Idle' states of the devices are predetermined states decided by the following logic: if the device's thermal behavior influences its performance, the device is left to operate to maintain its thermal stability. If it does not influence performance, the device is set to an 'off' state. For example, in the case of most RF amplifiers, when their gain is set by an analog signal, their amplification values may vary over time due to thermal changes influencing the properties of their circuit elements. This behavior can significantly impact experiments because the gain value will vary from sequence to sequence until new thermal stability is achieved. Therefore, the gain value in the 'idle' state is set to a fixed parameter. At the same time, the amplitude of the signal

from DDS sources is tuned down, as the DDSs have minimal variation in their performance over thermal stability. Another approach to address this issue could be running 10-15 dummy sequences before the actual ones, thus allowing the devices to gain thermal stability. However, this is rather unfavored since it would require specifying the dummy measurements by indicating them in the metadata of each readout.

A similar process is already carried out within every experimental sequence. A preliminary recording, known as a dummy frame, is captured by the CMOS cameras at 20 ms before acquiring the initial image in the fluorescence imaging phase. This procedural step facilitates the discharge of the pixel array in the CMOS camera, effectively minimizing noise during subsequent image capture. During this experimental sequence, the magnetic field gradient was not utilized to prevent atoms from being cooled down and loaded into the trap from the hot background gas during the imaging phase, as it would influence the determination of the final atom number of the sample.

Fluorescence readouts are not only conducted via CMOS cameras but also with photodiodes. Remarkably, employing photodiodes enables real-time fluorescence tracking, taking advantage of the fast Analog-to-Digital Converters (ADCs) available in our setup. In our case, the oscilloscope functions as an ADC. This differs from a CMOS camera, where the procedure entails saving an image and summing all pixel counts, underscoring the efficiency and speed inherent in our chosen approach. We have installed a mirror with a flippable base in the horizontal path, enabling the light to be directed either to the photodiodes or to the horizontal camera, which disallows the use of both devices simultaneously. A separate optical setup was not installed for the photodiodes due to limited optical access around the experimental chamber. If the photodiodes on the horizontal path are in use, the CMOS cameras in the vertical path can still be used for atom number or spatial distribution determination.

The measurements using photodiodes are detailed in the next section ([Section 4.4.3](#)), where their readouts are utilized with a lock-in detection tool for both cesium and potassium atoms to enhance the signal-to-noise ratio.

4.4.3 Lock-in detection of multi-species

The multi-species lock-in detection tool has been integrated into our fluorescence imaging setup, allowing users to measure fluorescence readouts with a higher signal-to-noise ratio. Consequently, it eliminates the impact of potentially problematic light noise sources, especially high-power lasers utilized around the imaging setup. This is particularly crucial in scenarios where the noise levels of these lasers might fluctuate during an ongoing experimental sequence, such as when shutters block or allow light to pass through, leading to light scattering and variations in the noise level within the medium.

The lock-in detection process effectively rejects noise at frequencies other than the modulation frequency (or its harmonics). Since the environment is not prone to introducing fluctuations at several kHz used for modulation, the light detected by the photodiode in a fluctuating background is rejected by the lock-in detection process.

The lock-in amplifiers require a reference signal, which can be expressed as $V_{ref} \sin(\omega_{ref}t + \theta_{ref})$, where ω_{ref} is the frequency and θ_{ref} is the phase of the signal. When the measured signal is given

as a $V_{sig} \sin(\omega_{sig}t + \theta_{sig})$, the lock-in amplifier multiplies these two signals via a phase-sensitive detector as can be expressed as:

$$V_m = \frac{1}{2}V_{ref}V_{sig} \cos([\omega_{sig} - \omega_{ref}]t + \theta_{sig} - \theta_{ref}) - \frac{1}{2}V_{ref}V_{sig} \cos([\omega_{sig} + \omega_{ref}]t + \theta_{sig} + \theta_{ref}). \quad (4.27)$$

When the signal V_m passes through a low-pass filter, it eliminates all the AC components, thus blocking the output signal apart from cases where the frequency of the input signal is equal to the reference signal frequency ($\omega_{sig} = \omega_{ref}$). In this particular case, the output would be only a DC signal with a phase difference dependence between the reference signal and the input signal. The corresponding DC output is

$$V_{out} = \frac{1}{2}V_{sig}V_{ref} \cos(\theta_{sig} + \theta_{ref}). \quad (4.28)$$

In real-world scenarios, the input signal often comprises a mixture of signals with different frequencies from various sources. These noise sources present challenges when directly reading photodiode outputs in a noisy medium, creating difficulty in extracting clear signals with a high signal-to-noise ratio. To address this issue, we have implemented a lock-in detection scheme for simultaneous use with both cesium and potassium atoms. The following section offers a detailed explanation of the design and functionality of the installed lock-in readout scheme within our single-chamber apparatus for KCs.

For lock-in detection to be effective, it is essential to have a controlled modulation of the fluorescence light emitted from the atoms. In our case, the fluorescence signal emitted from the cesium atoms is already modulated, a prerequisite for the chosen laser stabilization method implemented on those lasers as detailed in [Section 4.2.1](#). As the frequency of the cesium lasers is modulated, the fluorescence signal emitted from the ultracold sample also becomes modulated, as the scattering rate and resulting intensity of the fluorescence signal depend on the detuning of the laser (see [Equation 4.25](#)). A numerical determination of a lock-in-based fluorescence measurement is shown in [Figure 4.36](#).

The modulation frequency of our cesium cooling lasers is set to be around 46 kHz. As the laser diode current cycles up and down at a frequency of 46 kHz, its amplitude reflecting the change in the light's frequency causes a broadening of the laser's linewidth, which is approximately 4 MHz. The diode current modulation signal can be extracted from the laser control module, namely the '*DigiLock110: Digital Locking Module*' by Toptica Photonics. This signal is then amplified and directed to a lock-in amplifier (*SR830 - Stanford Research*) to be taken as an external reference modulation frequency for lock-in measurement. This signal is, in principle, sufficient for lock-in based fluorescence signal measurements. Nevertheless, it is necessary to introduce a modulation on the laser frequency for lasers without inherent modulation (such as our potassium lasers). This can be achieved by modulating the driving frequency of the AOMs used for the generation of cooling beams in our MOT, in which they are utilized for fluorescence measurements (which is detailed previously in [Section 4.2.1](#)). Moreover, the same approach can be used for our cesium cooling laser setup to increase its frequency-deviation to further increase the signal-to-noise ratio of lock-in readouts or even demodulate the existing modulation on its frequency.

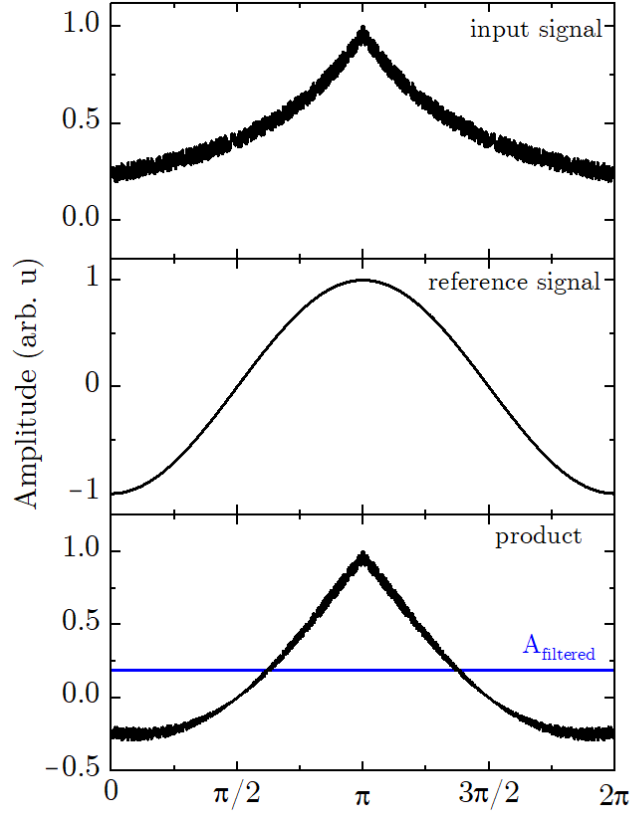


Figure 4.36: Numerical determination for a lock-in amplifier where a full-cycle readout of a fluorescence signal obtained from the MOT by modulating the cooling beam frequency by $\pm\Gamma$ centered around $\Delta = -2.29\Gamma$ from the atomic resonance (top). The lock-in amplifier generates the reference signal, which drives the frequency modulation of the cooling beam (middle). The product of the input and the reference signal, together with the value after its AC components, are filtered (bottom).

To control the frequency deviation of the cooling-MOT beams of both species, we have followed this procedure: The generated reference modulation frequency of the lock-in amplifier (synchronized with the ECDL modulation signal) is sent to an arbitrary waveform generator, which is configured to work on frequency modulation (FM) mode. This arbitrary waveform generator is utilized to generate a modulated RF frequency to drive the AOM facilitated in the generation of cooling-MOT beams. By doing so, the frequencies of the cooling-MOT beams of both species are modulated, and the resulting modulation is then used to detect the fluorescence signal via our lock-in amplifier. The schematic of the entire setup can be seen in [Figure 4.37](#). In the case of cesium lasers, demodulation of the light frequency is also achievable, as the AOM can be driven with an RF signal to compensate for the modulation introduced by the utilized stabilization method in cesium cooling lasers.

However, now that the cooling-MOT beams are modulated via an arbitrary waveform generator, it becomes necessary to integrate the waveform generator into our experimental control software.

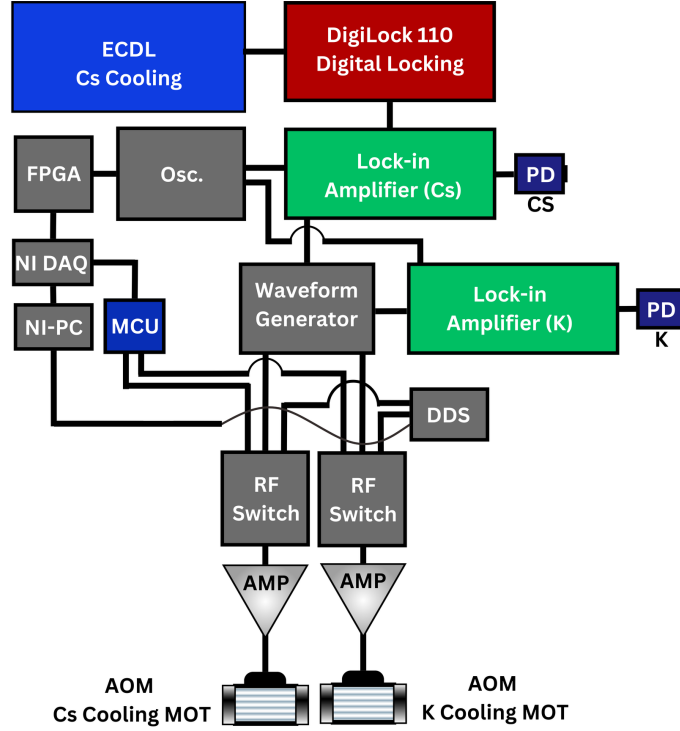


Figure 4.37: Schematic of the multi-species lock-in detection setup for cesium and potassium atoms. Key components include ECDL (External-Cavity Diode Laser), Osc. (Oscilloscope), MCU (Microcontroller Unit, Arduino Uno), and PD (Photodiode). Further details about the remaining devices, including FPGA (Field Programmable Gate Arrays, Opal Kelly XEM3001) and NI DAQ (National Instruments Data Acquisition), are provided in [Section 4.5](#). The optical components depicted in this figure are licensed under Ref. [\[121\]](#).

The AOMs responsible for preparing cooling-MOT beams are often used to generate other beams in various cooling and control phases of the experimental sequence. The integration of the arbitrary waveform generator into our experimental control software has become less practical since the remaining AOMs in our group are driven by DDSs, which already have a framework for seamless integration with our control software. Besides, our DDSs have predetermined operation modes in which their amplitude and frequencies can be changed with analog signals sent to the corresponding port [\[120\]](#), a feature that most waveform generators do not have. Thus, we have incorporated a controllable RF signal switch into our setup. This switch allows one to select the preferred RF source for driving the cooling AOMs used in our setup. By doing so, users can now apply a digital voltage (0/3.3 V) to a microcontroller (Arduino Uno), which has been programmed to facilitate the states of the RF switches. Consequently, the modulation (via arbitrary waveform generator) can be activated or deactivated (AOMs driven by DDS) based on the requirements during different stages of the experimental sequence.

The optimum frequency deviation of the cooling-MOT beam is determined by acquiring fluorescence readouts from a steady-state MOT. Firstly, the readouts' signal-to-noise ratios (SNR) are obtained using the internal modulation frequency of the lock-in amplifier, where a frequency

deviation of ± 2 MHz was utilized via the AOM. SNR is defined as the mean divided by the standard deviation. These measurements are shown in Figure 4.38c with a significant down peak in the SNR values around 22 kHz and 46 kHz.

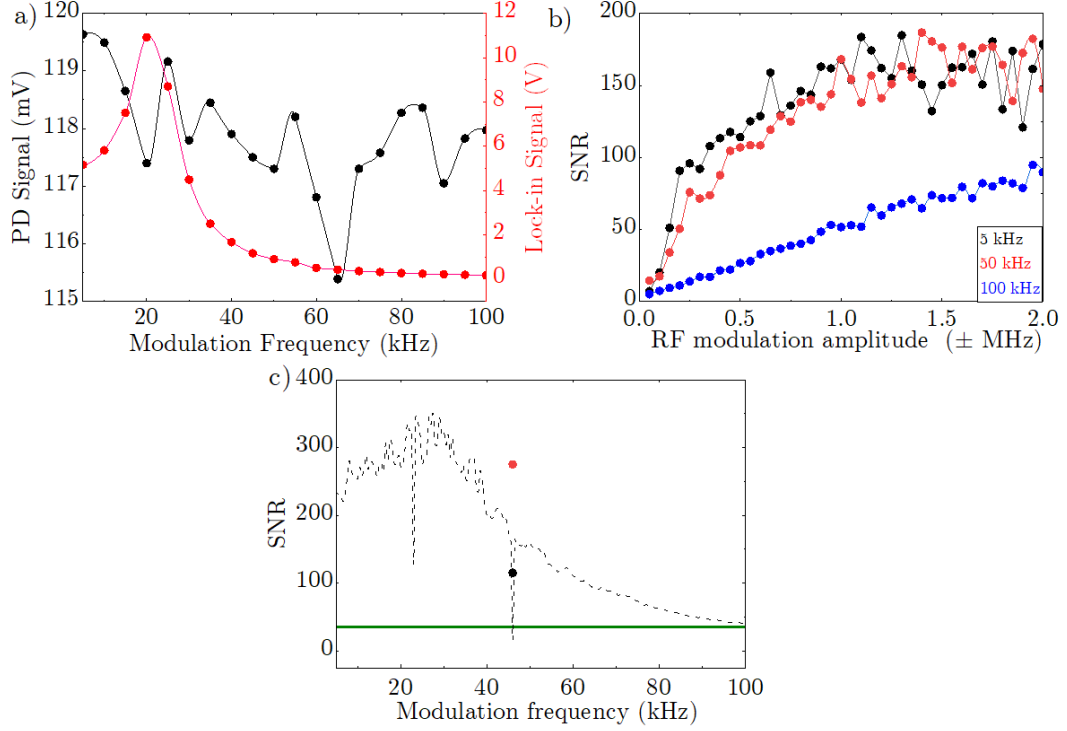


Figure 4.38: a) Fluorescence readouts of raw PD signal and lock-in amplifier signal are given as a function of the modulation frequency. b) The signal-to-noise ratio of the fluorescence readouts conducted with lock-in detection setup versus the frequency modulation (FM) introduced to the cooling MOT beam's frequency via an AOM. c) The signal-to-noise ratio versus modulation frequency of the lock-in detection setup, when the modulation frequency of the lock-in amplifier is set internally out-of-sync with the laser modulation (black dashed line), when they are synchronized (black point) and when they are synchronized together with the 2 MHz frequency deviation driven via an AOM. The green line indicates the signal-to-noise ratio of a raw photodiode output.

This phenomenon is attributed to the inherent modulation of our cesium laser, which operates at a frequency of approximately 46 kHz, with its phase being out of sync with the frequency deviation introduced with the AOM. We then set the lock-in amplifier to use the ECDL modulation frequency to be taken as the external modulation source, thus synchronizing the laser modulation frequency with the reference modulation of the lock-in amplifier. By doing so, the SNR was enhanced to approximately twice the SNR obtained with raw photodiode readouts when the additional frequency deviation was not used and eight times when it was utilized.

We have also investigated the relationship between the amplitude of the frequency deviation and the SNR. The frequency deviation of the RF frequency driving the AOM was varied between 0 to ± 2 MHz to determine the SNR under three different modulation frequencies when out of sync

with the cooling laser modulation. The results show a saturation of the SNR below ± 2 MHz for the frequency modulations $f_{mod} \leq 50$ kHz (see Figure 4.38).

This behavior is expected when the frequency of the cooling MOT beam is deviated excessively. Once the frequency deviation of the cooling beam is larger, the detuning of the cooling beam becomes closer and further from the optimum detuning for cooling the cesium atoms, thus leading to a reduction of the loading rate and consequently reduction of the detected fluorescence signal. In this context, the change in the atom number for an MOT can be given as the following [180]:

$$\frac{dN}{dt} = \alpha P_{Cs} - \gamma N - \frac{\beta}{V} N^2, \quad (4.29)$$

where N is the atom count, α is proportional to the trapping cross-section and P_{Cs} is the partial cesium pressure. γ is the one-body loss coefficient for collisions with the residual background gas, and β is the two-body loss coefficient. V is the effective volume in which, if the cloud has a Gaussian profile, it can be expressed as $V = (4\pi)^3 \sigma$ where σ is the width of the Gaussian cloud.

As the detuning results in a reduction of the loading rate at the same time, the rate of photons emitted per atom is enhanced since the increase in the fluorescence rate for lower detunings is higher than the reduction of the fluorescence signal at higher detunings (see the derivative of the fluorescence signal versus detuning of the laser as shown in Figure 4.39).

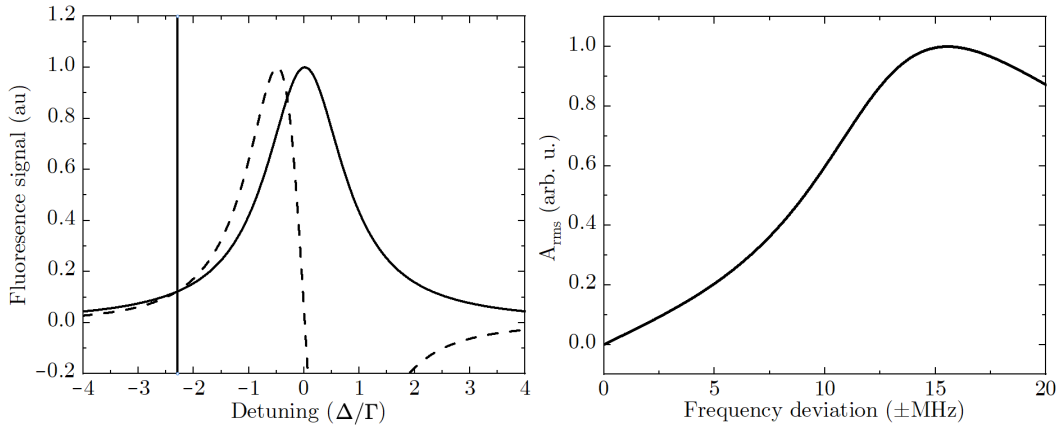


Figure 4.39: The plot on the left shows the normalized fluorescence signal rate versus the detuning of the cooling laser beam (solid line), along with its derivative (dashed line). On the right side, the numerical determination of the root-mean-square amplitude is presented in terms of the deviation of the frequency of the cooling beam centered around $\Delta = 2.29\Gamma$.

Thus, as the cloud loses atoms due to the inefficient cooling process, the fluorescence rate is enhanced. The ± 2 MHz of frequency deviation (when the modulation frequency is synchronized with the modulation of ECDL) gives the highest SNR value without influencing the atom count significantly. As the modulation frequency decreases, the steady-state atom number will be subject to fluctuations due to the loss of atoms from the trap following a loading. Since slower

modulation frequencies will give MOT more time to reach its maximum loading capacity, it will also have more time to lose atoms due to the decreasing loading rate.

In higher modulation frequencies, the atom count fluctuation is relatively lower compared to lower modulation frequencies since, in this case, there will be less time for the atoms to leave the trap due to heating before the optimized cooling process starts again. Thus, the steady-state atom count in a given laser detuning at time t_0 will never be approximated to the steady MOT loading rate given for the laser detuning at time t_0 .

In determining the measurements for the optimal SNR for our MOT, the modulation frequency was synchronized with the ECDL modulation, and the frequency deviation of the driving frequency of the AOM was chosen to be ± 2 MHz.

Our photodiodes are typically employed for measuring the MOT loading phase as shown in [Figure 4.40](#). Having such a setup offers an additional advantage—conditional MOT loading queries can be integrated into the experimental sequences. The primary goal of implementing this setup was to improve the consistency of our MOT loading phase. In our experimental setup, nearly all experiment sequences start with the MOT loading phase, followed by additional cooling techniques such as GMC. Consequently, the number of atoms loaded into the MOT becomes a key factor in obtaining consistent experimental results for each sequence.

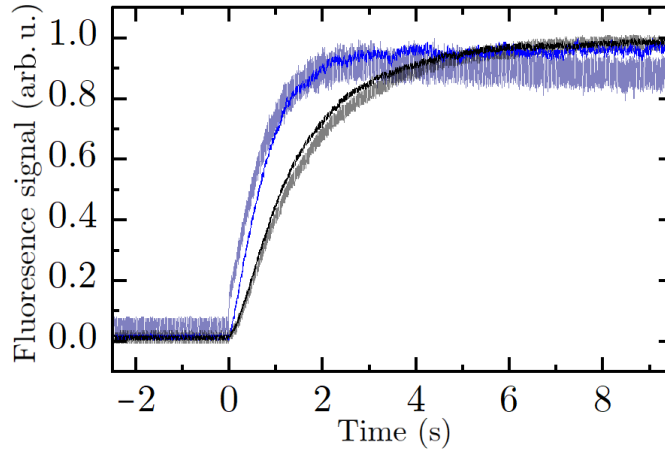


Figure 4.40: The MOT loading curves of cesium (black lines) and potassium (blue lines) were recorded via direct photodiode readout (dimmer lines) and lock-in readout (brighter lines). The lock-in readouts and raw photo-diode measurements were acquired at distinct time points, emphasizing variations in the MOT loading conditions.

When the MOT is loaded within a fixed time duration (typically 5 to 7 seconds), the number of atoms loaded into the MOT varies over time due to the fluctuating initial conditions of the vacuum chamber at the commencement of each iteration, such as the pressure of the background gas as expressed in [Equation 4.29](#). This variability can be detrimental to photoassociation measurements with a trap-loss-based readout. In such cases, the initial number of atoms loaded into the dipole trap can fluctuate from shot to shot. Consequently, weak photoassociation lines may be obscured in the fluctuating baseline of the remaining atom count. To address this issue, we utilize photodiodes to monitor the MOT loading phase, with the option to send trigger signals

to the experimental control software at user-specified fluorescence levels, – instead of relying on a fixed loading time as a proxy for atom number. This approach ensures more repeatable initial conditions for atoms in the dipole trap. Coupled with lock-in detection, the minimization of fluorescence fluctuations further enhances the reliability. The conditional MOT loading is discussed in [Section 4.5.2](#).

Moreover, with the implemented lock-in detection scheme, one can retrieve MOT loading curves with minimized noise. The loading curve of individual species can be analyzed to retrieve the pressure in the chamber and the collisional dynamics of ultracold atoms and hot residual gas by using the expression given in [Equation 4.29](#) [180]. Since our samples are designed to cool down from the hot background gas, the determination of background pressure under varying conditions plays a crucial role in determining optimum parameters, such as how much current should pass through the dispensers and what should be the optimal pulse duration of the LIAD (light-induced atomic desorption). LIAD is a technique employed to desorb atoms from the walls of the vacuum chamber by exposing a broadband UV light, rapidly increasing the pressure within a short duration. This facilitates achieving lower background pressure and fewer background collisions in the later stages of the experiment. It is crucial to utilize the LIAD in the right way in the single-chamber setups where the lifetime of prepared samples in magnetic traps and optical dipole traps can be extended further.

The impact of LIAD usage in our setup was investigated by doctoral student Jakup Dobosz [181], utilizing the implemented lock-in detection scheme. In the initial phase of the experiment, the relationship between the MOT loading rate and the steady-state number of atoms was studied with varying depending on the given LIAD current and, thus, the intensity of the UV light. Briefly, the MOT was let to reach a steady state following a LIAD pulse for 10 seconds of fixed duration. By applying the analytical solution of the MOT loading equation to both segments and extracting the steady-state number of atoms, the parameters in the loading rate equation ([Equation 4.29](#)) were determined. The results indicated that the LIAD current was initially used excessively; thus, its typical operation current was reduced to half of the initial value. The second measurement involved MOT loading with LIAD, which was activated initially and then deactivated. In the first part, the pressure, enhanced by LIAD, is assumed to be constant. In the second part, a characteristic exponential decay is observed. This behavior is anticipated, as the velocity of gas flow through an aperture is proportional to the pressure difference. The results from both measurements indicate that upon turning off LIAD, the background pressure undergoes a substantial drop, approximately 40 times.

Furthermore, the lock-in detection scheme becomes helpful for extracting collisional properties between different species from their individual and simultaneous loading curves. While research in this context is available for other species such as Rb-Cs[182], K-Rb [183] and Li-Rb[184], it has not been yet done for K-Cs atoms in depth. This implemented lock-in detection scheme can be employed to evaluate collisions between potassium isotopes and cesium atoms in the MOT, providing less noisy readouts.

The change in the number of atoms of one species when two are present in the MOT can be expressed as the following [184]:

$$\frac{dN_A}{dt} = L_A - \gamma_A N_A - \beta_A \int n_A^2 d^3r - \beta_{A,B} \int n_A n_B d^3r, \quad (4.30)$$

where, N_A is the number of atoms in the species A MOT, n_A and n_B are densities of MOTs of species A and B, respectively, and L_A is the loading rate for species A. γ_A is the one-body loss rate coefficient accounting for the losses due to collisions with the background gases, β_A is the two-body loss rate coefficient accounting for the losses of species A due to collisions between atoms of species A, and $\beta_{A,B}$ is the two-body loss rate coefficient accounting for the losses of species A due to collisions with species B. The ordering of indices in $\beta_{A,B}$ is significant, where the first index denotes the species undergoing losses, influenced by the presence of the species indicated by the second index.

These coefficients can be obtained by conducting four sets of measurements of the MOT loading curves for our cesium and potassium atoms. First, the cesium atoms are loaded into the MOT and tracked until the cesium sample reaches a steady state; this way, the parameters for the L_{Cs} and γ_{Cs} is determined. Then, the potassium atoms are loaded into the trap with an intentionally reduced loading rate. By doing so, the $\beta_{K,Cs}$ can be determined via the expression:

$$\begin{aligned}\beta_{Cs,K} &= (L_{Cs} - \kappa_{Cs}\bar{N}_{Cs}^\infty) / (\bar{n}_K\bar{N}_{Cs}^\infty), \\ \beta_{K,Cs} &= (L_K - \kappa_K\bar{N}_K^\infty) / (\bar{n}_{Cs}\bar{N}_K^\infty).\end{aligned}\tag{4.31}$$

Subsequently, the MOT is turned off for a sequence, during which the potassium is loaded into the MOT and tracked until it reaches its steady state, allowing us to determine L_K and γ_K , followed by loading of cesium atoms with reduced loading rate into the MOT and the $\beta_{Cs,K}$ is determined. These measurements have yet to be finalized. However, the initial preparation and the technique to be used are outlined here, as they are planned for the near future as part of ongoing research.

4.5 Experimental control

Experiments involving ultracold ensembles typically require precisely timed series of events with dynamically tuned parameters, such as operating frequencies and signal amplitudes, to meet the specific requirements of a given measurement. Executing these experimental procedures demands precise and reliable timing tools, especially considering the need for repetitive runs that must deliver experimental sequences in identical conditions. These demands are fulfilled via our experimental control hardware, which consists of three computers and various I/O devices connected to a main experimental control computer.

This section provides information on how our devices are controlled through analog and digital sources, local-area-network servers, and data management procedures for storing experimental readouts and conducting subsequent analyses. Moreover, the section also details the methods employed at the timebase of the experimental control hardware. Timebase here is defined as the time period used as a base for device control. This section also provides a comparison between fixed timebase and variable timebase. Notably, users can incorporate basic logical (AND/OR) operations into their experiments in a variable timebase. Beyond its efficient RAM usage for handling complex and lengthy experiment sequences, the variable timebase presents a crucial advantage by allowing users to modify time-varying processes that, under a fixed timebase, cannot be addressed with predetermined sequence timings.

Our group occasionally faces compatibility challenges between the control setup and experimental devices, especially when these devices lack a direct analog control port. While it is unusual for research-oriented devices to lead us to such issues, some devices, by their operation principle, necessitate predetermined settings rather than live control ability. For instance, devices like our DDSs only offer predetermined profiles in their settings. As a result, users need to configure these profiles before the experiments, and subsequently, these profiles can be adjusted via analog voltage sources. Similarly, the real-time tuning capability is primarily supported by most AWGs only within a specific operation mode. However, it's important to note that the operation mode of these AWGs cannot be altered via precisely timed analog signals.

The main controller of the experimental control setup is a computer by 'National Instruments' facilitating PXI (PCI eXtensions for Instrumentation, where PCI stands for Peripheral Component Interconnect) system. Multiple analog and digital PCI cards, guided by the main computer, act as sources for TTL and analog/digital signals in the experiment. Additionally, it manages external devices through HTTP, although this method is solely employed for transferring data and setting up the configurations of the files, which does not require precise timing. A second computer is exclusively dedicated to operating CMOS cameras and is utilized for data acquisition. In this setup, all other devices, if needed, would send metadata to this second computer. The readout will then contain the metadata received from all devices. The PXI system controls the parameters and triggers of these cameras. A third computer is tasked with handling post-experimental data processing duties.

4.5.1 Sequence control software

The sequence control system runs on two software components developed by Keshet, A. and Ketterle, W. [41] for atomic physics experiments. One of these components is Cicero, a GUI client that enables users to control digital and analog outputs and create predetermined experimental sequences essential for atomic physics experiments. The other component, Atticus, is a server responsible for configuring output hardware. Atticus transforms user inputs from Cicero sequences into an output buffer generated for the server computer's output hardware. Cicero and Atticus are coded in C#, and users can access their source code for modifications.

Significantly, these programs underwent some modifications [120] to address specific issues and improve user experience in sequence management. The modifications were introduced to resolve issues such as the lack of user-friendly features, the absence of unique sequence names, and the inability to save sequences with essential parameters.

The Atticus server functions as the central hub for controlling the settings of all available devices within the control hardware. Users can customize various parameters, including the device's time resolution, clock source, and the activation or deactivation of specific channels. Atticus facilitates user access to all configuration settings through a GUI, eliminating the need to alter hard-coded parameters within the source code. The general architecture of the control system (both hardware and software) is shown in [Figure 4.41](#).

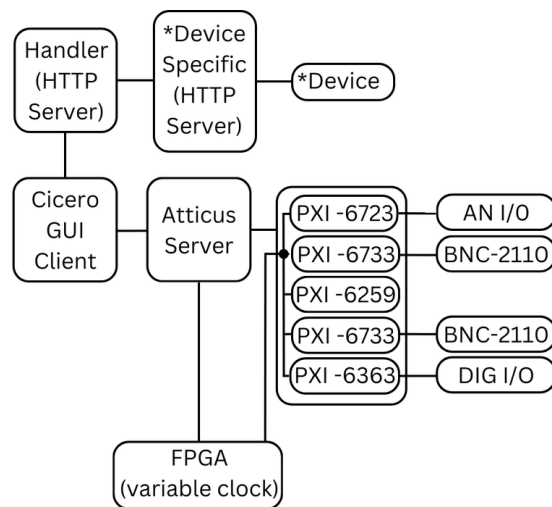


Figure 4.41: The architecture of the control system, including electronic components and software tools. Cicero, a GUI client for user input, and Atticus, a server responsible for managing analog and digital channels. Atticus is utilized to control the PXI cards connected to the modules where analog and digital input and outputs can be accessed. The FPGA is employed to distribute the variable clock source to PXI modules. The Handler, functioning as an HTTP-based server, also manages device-specific servers accessible through the TCP/IP communication protocol. PXI-6259 is currently on reserve for future usage.

Users can manage devices connected to the Atticus server on the Cicero GUI interface. These devices include PXI modules with analog and digital channels to which users can assign user-defined names in the GUI. For instance, linking the PXI-6733, designed for high-speed analog output in control and waveform applications, with the BNC-2110, a block module enabling access to analog and digital signals along with two user-defined connections, allows users to designate any port on the BNC-2110 module. For example, the designated ports can be renamed, providing a user-friendly approach to identifying the specific function of each port in the system. Once a port is assigned, users can easily incorporate timing blocks. The ports with user-specified names can be added within these blocks with a specified output value. Users are, in principle, allowed to insert an unlimited number of timing blocks, enabling sequential control of all available output ports. This process can be replicated for all other analog and output ports of PXI modules.

In the prepared sequences through the Cicero GUI, users can also send commands to other devices via a simple HTTP-based server designed to handle commands, depicted in the control software layout (Figure 4.41). This way, the devices that cannot be controlled via digital and analog signals but via LAN interface can be controlled via HTTP commands. Despite Cicero's inherent limitation in communicating directly via the HTTP protocol, the 'Handler'(developed using the Python programming language.) creates a virtual RS232 port that Cicero treats as genuine, receiving commands as if transmitted through RS232 communication. The 'Handler' actively "listens" to these commands and interprets their content. Subsequently, based on the command content, the Handler transmits the appropriate command via the LAN interface to the targeted devices. TCP/IP communication, when employed within the same local area network, typically requires approximately 2-3 milliseconds for data transmission. Devices receiving commands may exhibit varied response times when adjusting their settings based on the received commands.

Consequently, TCP/IP communication is not ideal for precise timing. Instead, these servers are frequently utilized to set a device's modes, especially at the start of each sequence, ensuring the device is in the correct mode before undertaking its corresponding task in the experimental sequence.

The servers discussed above make communication with peripheral devices relatively easy. In virtually every experiment, we rely on the dedicated servers designed for data readout from CMOS cameras, which are used to acquire images of the ultracold samples in our experiments. At the beginning of an experimental sequence, the device-specific server for CMOS cameras receives a 'start' command, indicating the initiation of an experiment. Upon receiving this command, the server retrieves the sequence name (user input from the Cicero GUI) to name the output files appropriately. Subsequently, parameters like exposure time can be sent to the server, which handles communication with the device it is responsible for via drivers provided by the manufacturer. Another critical feature is the ability to embed all necessary parameters into the metadata of the images captured during that sequence. This allows users to trace back all the data used in a specific sequence. The Cicero-Atticus software's source code theoretically permits imprinting the entire experimental sequence as metadata. At the end of the experimental sequence, the command 'end' is used to inform the camera server about the end of the experimental sequence. Then, the data stored in the camera's buffer is uploaded to the computer running the server. From there, the new data is copied to another computer and is solely used for data analysis and image processing. This happens at the end of each experimental sequence. With this approach, the operation of each data-gathering device is decoupled from data processing. As such, it makes the entire infrastructure safer because the failure of the computer that controls, for example, the CMOS camera, does not influence the computer running the experimental sequence or the one responsible for data storage and analysis.

4.5.2 Variable timebase

In this section, one can find integrating a variable timebase configuration into the Atticus/Cicero experimental control system introduced in [Section 4.5.1](#). Users of this setup can choose between a fixed timebase and a variable timebase. While both options share similarities in functionality, the focus here is on the unique features of utilizing the variable timebase. It is important to note that Cicero and Atticus are specifically designed to operate with National Instruments cards. By default, the entire system is configured to provide information about the settings of each available channel for every clock signal. However, this default configuration can lead to issues, such as extended computation times and high memory usage, mainly when dealing with long sequences of signals. Atticus and Cicero do not have the variable timebase clock implemented by default. While instructions are available on how to implement it, these instructions do not cover all extension cards, and there may be ambiguity regarding the procedure, especially with our specific cards. Despite the likelihood of similarities in the procedure across most cards, it is essential to outline the specific steps taken in this instance.

Utilizing the variable timebase configuration offers a considerable improvement over the shared-clock version, which uses a fixed timebase method. The shared clock is a signal source responsible for sending trigger signals at fixed intervals, whereas the variable timebase awaits the following change in the predetermined signal. Consequently, the sample size is significantly reduced, leading to efficient buffer generation. The sample size represents the designated memory space

where data points from analog and digital channels are stored over time. This allocation, also known as the buffer size, is crucial for accommodating the generation of these data values. This process may take several seconds, mainly when used with a shared clock. By transitioning to a variable-time base, our revised system operates differently from the previous one. In the former system (fixed timebase), when a user predefines an experimental sequence, the computer control system allocates a digital value in the memory for each time step of the sequence corresponding to each device integrated into the control system. This means that for a given clock rate (usually around or less than μs in steps), the control system generates values for every clock cycle for each device and then allocates them in memory. In contrast with having around 50 independent devices connected to the system and the usual sequence time being around 15 seconds, the control software occupies almost the entire memory of its available size. In addition, generating the sequence typically takes around 2-3 seconds, which becomes an obstacle for speeding up the experiments conducted with this system. It is also worth mentioning that once the sequence starts, the user cannot change any parameter or, most importantly, add a feedback control mechanism in the setup. However, in the variable timebase configuration, the device is triggered by an external clock source only when a change exists in any device included in the system. For instance, let us assume that the user sets a clock rate of 1 MHz. Then, it runs an MOT loading process, where all 50 devices used in this segment of the experimental sequence have fixed constant analog control signals for 6 seconds. The control system with this configuration will generate 300M data points only for this sequence segment. Assuming that the control system has a 64-bit setup, 2.4 GB of memory will be occupied only by this segment. In the variable timebase configuration, only one data point will be generated for each device. In the end, there will be only 400 bytes assigned to the memory. This makes a massive difference since one can increase the clock rate and have better time resolution of the analog and digital control channels over the experimental devices without worrying about the allocated memory size.

The variable timebase is regulated through a field programmable gate array (FPGA) Opal Kelly module (XEM3001v2), an external clock source for our setup. This FPGA has firmware that dictates the timings for holding or allowing the clock cycle to proceed. These timings are established by assessing the modifications to be implemented in the devices. The clock cycle is continuously employed whenever a change is initiated in any device. However, the clock cycle is halted during any time interval where each specified data point for all devices is identical—comparing two timebases regarding timeframe count in [Figure 4.42](#).

Our PXI cards can use one of their input channels configured as a trigger for executing additional operations. The input channel is usually an internal trigger channel where the clock signals are received directly from the shared computer between all devices. We rely on the Opal Kelly (XEM3001v2) module to provide this trigger. The Opal Kelly module (XEM3001v2) currently interfaces with two PXI modules, namely PXI-6733 and PXIe-6536.

PXIe-6536 exhibits an unfavoured behavior in which it is not designed to have direct synchronization with the internal clock-source channel (PXI-Trig0). Consequently, the PFI4 port of PXIe-6536 was configured to utilize the clock-cycle readouts obtained from the Opal Kelly module. PFI is "Programmable Function Interface," which refers to programmable digital I/O (Input/Output) lines. PFI lines can be configured for various purposes, such as digital input, digital output, or trigger lines for specific events. This unfavoured design is a result of the PXIe-6536's inability to perform counting operations for regular I/O channels, a necessary operation for reading the signals from shared channel PXI-Trig0. PXI-6733, on the other hand, successfully receives the signal from its PFI0 port, which is then internally routed to its PXI-Trig0 port. All other devices

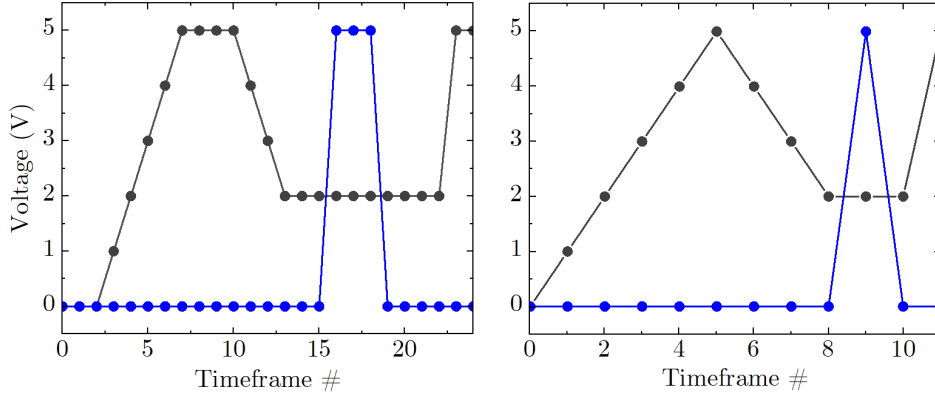


Figure 4.42: Two timebases are presented: a fixed timebase on the left and a variable timebase on the right. The data points for two channels, analog (in black) and digital (in blue), are provided concerning the timeframe count they utilize for the same experimental sequence.

connected to the same computer receive the clock signals from this shared PXI-Trig0 channel.

While integrating the Opal Kelly module into the Atticus Server, we encountered issues necessitating the rebuilding of Atticus in 32-bit to address a discrepancy related to 32-bit/64-bit installations on the computer. The firmware for Opal Kelly, intended for implementing variable timebases, is likely designed to operate with 32-bit builds and is not compatible with 64-bit builds. As explained in [Section 4.5.1](#), these software components are developed in the C# programming language. When modifications are necessary, the source code must be loaded into Microsoft's Visual Studio to be built with the desired settings, including the operating system's bit count. The need for rebuilding arises because once Visual Studio builds a project, it becomes standalone software, eliminating the need for additional package installations and mitigating potential issues that may arise over time due to package updates.

After the implementation, the timings of selected channels from four PXI cards were examined for synchronization, as illustrated in [Figure 4.43](#). To avoid possible time delays introduced by the cards' connections with the oscilloscope, we ensured that the BNC cables were of the same length (10 m). The observed offset difference between devices was less than 500 ns.

After implementing the variable timebase, we proceeded with the setup to enable conditional triggering. We use the conditional triggering scheme on the MOT loading phase of the cesium and potassium atoms. At the beginning of each experimental sequence in our single-chamber KCs, we initiate the loading of hot background gas into the MOT. Using fixed-time durations for the loading stage resulted in variations in the MOT loading times, leading to fluctuations in the initially prepared number of atoms for subsequent measurement stages. To minimize these fluctuations, we employ conditional triggering, enabling progression to the following experimental time frame only when a specific level of the fluorescence signal of the atoms in MOT is reached. The fluorescence signals are measured using a lock-in detection scheme.

To record variations in the prepared samples, we first load hot background gas into the MOT with a fixed time duration and then capture images of the atoms using a fluorescence imaging sequence. Subsequently, we employ conditional triggering, allowing the MOT to load until a

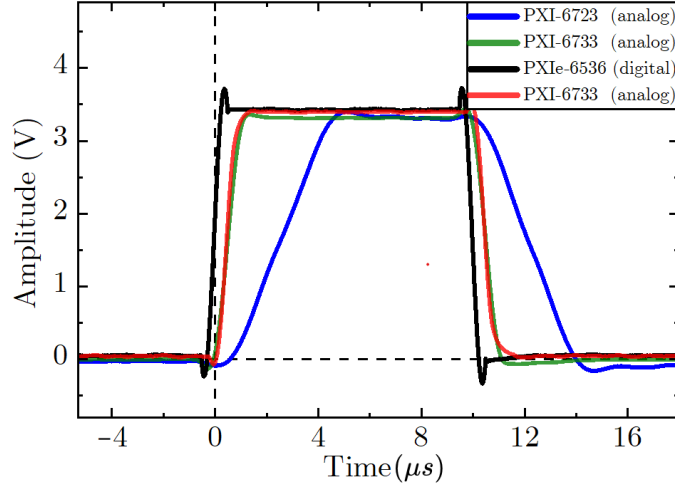


Figure 4.43: Examples of selected output ports for every PXI module (PXI-6723, two PXI-6733, and PXIe-6536) currently operational in the experiment control scheme. The same clock pulse triggers all four channels (three analog and one digital) simultaneously to show their synchronization with the external clock source.

specified fluorescence signal level is reached. We then calculate the coefficient of variation for both scenarios, which is defined as the ratio of the standard deviation to the mean.

Upon comparing both cases, we observe a significant reduction in the coefficient of variation by a factor of 4. Without conditional triggering, the coefficient of variation can reach as high as 3.09% within a hundred iterations of the experiment. However, when activated, it can be decreased to 0.75%.

This demonstrates a substantial improvement in readout consistency and the reduction of fluctuating trends caused by the changing conditions of our single chamber. The reasons behind these fluctuating trends vary; however, we can name several. In Figure 4.44, the grey and red data points represent the readouts corresponding to a fixed time duration for loading ultracold cesium atoms into the MOT. The initial data points in these measurements show an increasing trend, indicating that, within a fixed time for MOT loading, more atoms are loaded in each iteration until it reaches a stabilized value.

This behavior was investigated by monitoring the laser intensities of the MOT beams through iterative sequences. The cooling and repumper beams of the MOT are generated by altering their frequencies using AOMs. The RF signals for driving the AOMs are produced by a DDS or AWG and then amplified to the desired power using amplifiers with amplification ratios controlled by analog voltage signals. Initially, the RF amplifiers are set to have a gain value of 0 during idle periods. Consequently, these RF amplifiers supplying AOMs exhibit lower temperatures. We have determined that thermal fluctuations influence the gain of the RF amplifiers. Thus, when the experimental sequence is executed iteratively, the RF amplifiers reach a new thermal stabilization point. Until thermal stabilization is reached, the resulting atom count leads to inconsistent MOT beam powers for the first few iterations of each experimental sequence.

Regarding the observed downward trend in the grey data points in Figure 4.44, we attribute it to the decrease in LIAD efficiency. LIAD (Laser-Induced Atom Desorption) is a technique used to free atoms from the walls of the vacuum chamber by exposing them to broad-spectrum UV light. When the atoms are released from the surfaces, a sudden increase in pressure is observed within a short duration.

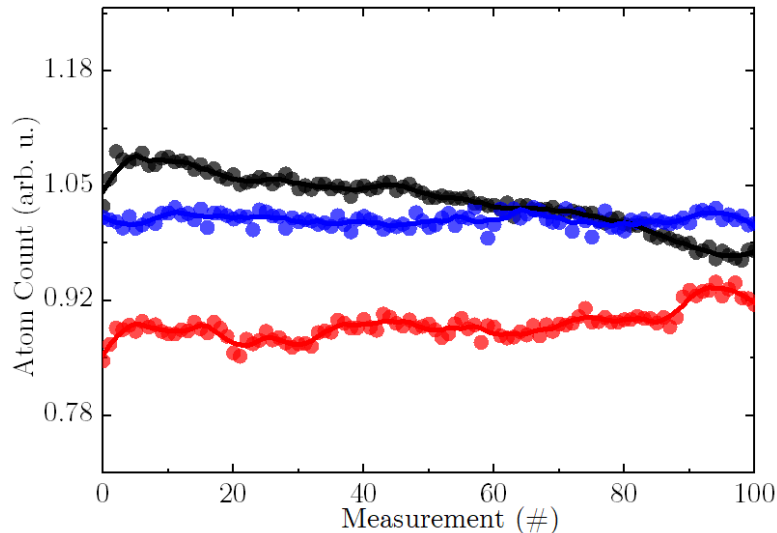


Figure 4.44: The graph presents three datasets, each acquired under distinct loading conditions. The gray and red datasets were captured with a fixed loading time for the Magneto-Optical Trap (MOT). The gray data points collected initially exhibit a downward trend indicative of the diminishing effectiveness of Light-Induced Atom Desorption (LIAD) over time. In contrast, the red data points, acquired after repeated LIAD applications, represent the behavior after reaching a steady state. The blue data points were obtained with a variable timebase together with the usage of conditional triggering.

The primary goal of LIAD is to attain a higher background pressure during the MOT loading stage and a lower background pressure to reduce the occurrence of background collisions in the subsequent stages of the experiment. When the experiments are run iteratively for an extended time, the efficiency of the LIAD remains constant. The effective LIAD efficiency might be associated with the high pumping speed of the ion pump: the UV light efficiently removes atoms from the inner walls of the glass cell, and they are subsequently pumped out from the trapping region. Consequently, the alkali atom layer deposited on the walls decreases.

4.6 Photoassociation lasers

This section details the optical setups designed for conducting photoassociation spectroscopy of ultracold atomic samples of cesium, potassium isotopes, and cesium-potassium mixtures. Additionally, the constructed setup was designed in consideration of the need to develop a laser system for single-color photoassociation for probing vibrational levels in the excited states of molecules [42] and two-color photoassociation for probing ground-state vibrational levels [185, 186, 187] and

Stimulated Raman Adiabatic Passage (STIRAP) [17, 118] for the formation of dense ultracold ground-state KCs molecules. As the constructed setup is designed to have features that allow us to conduct experiments on the above-mentioned schemes, the setups in this work were only used for single-color photoassociation of ultracold cesium atoms.

In a single-color photoassociation scheme, as previously discussed in Section 3.1.2, two colliding atoms absorb a photon to form an electronically excited molecule. Single-color photoassociation is utilized via a single laser that is tuned to a transition that couples the energy of two free atoms into an energy level at an excited molecular energy level.

Moving forward, two-color photoassociation involves the use of two tunable lasers. The first laser is employed in the single-color photoassociation, where it couples the transition between two free atoms and a weakly bound electronically excited molecular state. This introduces atom loss in the prepared ultracold sample since it leads to the formation of molecules. Then, a second laser is employed in this scheme, and its frequency is scanned around the frequency of the transition between the weakly bound excited state and a ground molecular state. When the frequency of the second laser beam becomes resonant with the transition coupling the excited and ground molecular states, it leads to a reduction in the rate of free-bound photoassociation, forming an Autler-Townes doublet [185, 186]. The Autler-Townes doublet is a phenomenon where a transition splits into a doublet due to the dynamic Stark effect of an oscillating electric field. The introduced loss of molecular formation rate is then used as a function to probe the ground state vibrational levels [188, 189]. Once the ground state vibrational levels are determined, these ground state levels can then be populated via the STIRAP scheme.

STIRAP is a laser-based method that allows efficient and selective transfer of population between molecular states. This process involves at least two coherent light sources [95]. The lasers here need to have extremely low relative short-term phase jitter and high long-term frequency stability to allow for coherence. The first one, often referred to as the "pump" field, is used to excite the system from an initial state (a pair of free atoms) to an intermediate state (a molecular state in an electronically excited potential). The second one is referred to as the "Stokes" field, which is then used to facilitate the system's transition from the intermediate state to the final state (a bound level in the electronic ground state potential of the molecule). The sequence of these transitions is controlled in the following way with short light pulses (typically a few tens of μs) of the above-mentioned transitions. The Stokes field is driven before the pump transition, which ensures that the population is transferred from the initial state to the final state without populating the intermediate state. Specifically for dimers, STIRAP has been used on atoms that have been laser-cooled and brought together to form molecules through magnetoassociation [190] or photoassociation [102]. Once these weakly bound molecules are created, they can be efficiently manipulated to ground state using STIRAP with exceptional efficiency above 80% [118]. This exceptional efficiency allows one to even achieve molecules populated in the rovibronic ground state via four-photon STIRAP schemes with resulting above 50% efficiency [17].

Let us now consider the wavelength range of tunable lasers. One should be able to conduct single-color photoassociation of cesium and potassium isotopes, as well as two-color photoassociation and STIRAP for cesium-potassium mixtures. The single-color photoassociation of cesium atoms requires laser beams tuned above 852 nm for the molecular states dissociating towards $6S_{1/2} + 6P_{3/2}$ asymptote and above 894 nm for the molecular states dissociating towards $6S_{1/2} + 6P_{1/2}$ asymptote. For the single-color photoassociation of potassium atoms, it is required to have wavelengths above 767 nm for the molecular states dissociating towards $6S_{1/2} + 6P_{3/2}$

asymptote and above 770 nm for the molecular states dissociating towards $6S_{1/2} + 6P_{1/2}$ asymptote. Whereas for the potassium-cesium mixtures, our intention is, as stated above, to have the formation of ground state ^{39}KCs molecules. A STIRAP scheme for ^{39}KCs molecules was previously reported in a theoretical work by Borsalino et al. [103]. In their study, the authors reported multiple transitions for both the pump and Stokes fields, aligning with wavelengths spanning from 674 to 712 nm and 925 to 999 nm, respectively. Our titanium sapphire lasers, including the SolsTiS-SRX manufactured by MSquared Lasers and the ‘TIS-SF-777’ manufactured by Tekhnoscan Laser Systems, cover this wavelength range up to 955 nm. With their collective tunability ranging from 650 nm to 955 nm, these lasers are well-suited for our purposes. This wavelength range adequately accommodates all current and future planned projects in this context (see Figure 4.45).

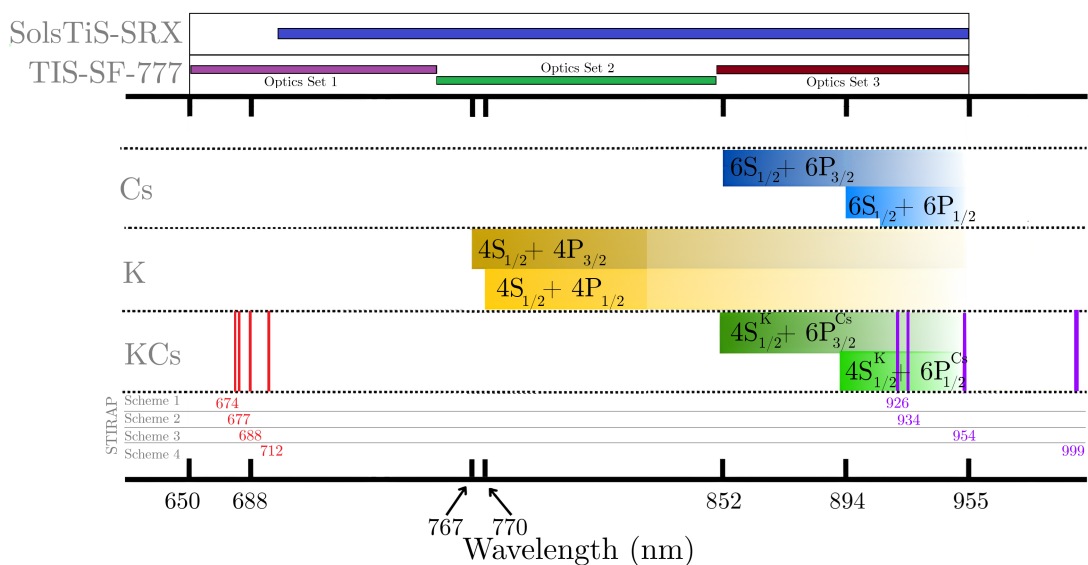


Figure 4.45: The wavelength range for the titanium-sapphire lasers SolsTiS-SRX and TIS-SF-777, along with the regions where single-color photoassociation spectroscopy of Cs_2 , K_2 , and KCs , can be conducted. The violet and the red vertical lines shown at the bottom of the figure mark laser wavelengths that have been determined by Borsalino et al. [103] as optimal for the production of ^{39}KCs ground state molecules by via STIRAP.

The SolsTiS-SRX has high efficiency in terms of output power while enabling users to conduct larger frequency scans up to 25 GHz while locked to its external reference cavity. Additionally, its wavelength can be tuned within the range of 720 nm to 950 nm without the need for any modifications to the elements in the laser cavity, making it ideal for probing the vibrational energies of weakly bound K_2 , Cs_2 , and KCs molecules in the excited electronic state. Therefore, we designed the SolsTiS-SRX laser to primarily utilize the single-color photoassociation experiments with the intention of probing vibrational levels in the excited molecular states. Additionally, it can also serve as a Stokes laser for the proposed STIRAP scheme for ^{39}KCs molecules.

In contrast, our second laser, the ‘TIS-SF-777’, features three different optical element sets covering various spectral ranges, necessitating significant effort and realignment of its cavity elements

with each change of optics. Additionally, it only allows for a smooth scanning range of 4.8 GHz, relatively narrower than our primary photoassociation laser. Therefore, this laser is assembled specifically for use in the STIRAP process after determining the excited-state vibrational levels with the SolsTiS-SRX. Consequently, it can be used as a pump laser covering the range around 688 nm in the planned STIRAP scheme for ^{39}KCs molecules to efficiently transfer the population from the excited state to the ground state. The installation of the aforementioned two titanium sapphire lasers in our setup has been a part of the development of the infrastructure. The initial testing done with Cs_2 molecules turned out to open possibilities to focus on novel measurements only in these homonuclear species. Nevertheless, the developments described in this chapter have been essential steps towards experiments with heteronuclear species.

Even though the lasers we use are fairly standard commercial products, their installation is not straightforward and requires months of alignment, optimization, and fixing unexpected technical problems like broken piezoelectric transducers (PZTs) or frequent misalignment of the laser cavity due to the drift of intra-cavity mirror mounts. These issues have been solved, as discussed in the following sections, but they caused significant delays in the planned research program.

Both titanium sapphire lasers we have installed are optically pumped by a single 532 nm, 15 W Lighthouse Photonics Sprout laser. Unfortunately, this laser was also another source of issues manifested as low output power or abrupt power drops, which would make it impossible to operate both titanium sapphire lasers simultaneously. Thus, we have opted to reinstall the laser setup following the user's manual to ensure no crucial step influencing the final performance was missed.

The re-installation began by addressing the discrepancies mentioned above to ensure the effective operation of the pump laser. The following sections provide a detailed account of our efforts to overcome these challenges and prepare our laser systems for the upcoming experiments, starting with the pump laser and proceeding to the titanium sapphire lasers. In the following section, one can also find details of the light distribution systems for each titanium sapphire laser that consists of several optical paths. One path, where an AOM is used to provide fine control of power sent to the atoms and to quickly turn off the laser light, is coupled into a polarization-maintaining fiber used to deliver light toward the prepared ultracold atomic ensembles. Another path with a wavemeter is used for frequency readout. The final path is used for obtaining a beat note with a frequency comb, which enables referencing the lasers to an optical frequency comb, which is detailed in [Section 4.6.2](#).

The assembly, maintenance, and optical referencing of the aforementioned lasers have been performed solely by me.

4.6.1 Installation of two titanium sapphire lasers

The installation process began with relocating the lasers to the same optical table where the vacuum system was installed and reinstalling our pump laser Sprout-G laser. Our pump laser Sprout-G comprises two main parts: a primary module housing high-power laser diodes, cooling systems, and the control interface, and a second module (laser head) containing an SHG crystal and an output window. SHG crystal refers to the second harmonic generation crystal. Second Harmonic Generation is a nonlinear optical process in which a crystal is used to generate light at twice the frequency (half the wavelength) of the input laser light. The laser head module

and the primary module are connected by a cable conduit housing cooling water carriers and a polarization-maintaining high-power optical fiber. Unfortunately, the cable conduit had experienced stress and bends in the past, leading to issues with the optical fiber connecting the two modules. When the pump laser was activated, it only reached 12 W of power before rapidly decreasing to below 2 W and then attempting to ramp its power again. Despite several attempts, the laser entered maintenance mode without resolving the power issue. Suspecting damage to the optical fiber due to bending, we decided to change the orientation of the cable conduit by applying gentle force. It was discovered that the maximum available power the pump laser could provide was related to the orientation of the cable conduit. After trying hundreds of different orientations, we achieved the promised maximum power of 15.25 W by the manufacturer. The cable conduit was then secured to maintain its orientation. Crucially, the cable conduit had to be straightened at the connection ports to both modules for at least 15 cm. Once the laser head and module were fixed in their desired locations, we performed general maintenance on the laser by connecting to the laser module via serial connection. Using the *‘LighHouse-RC’* software provided by the manufacturer, I initiated maintenance for maximum power and low-noise settings. This hour-long process involved the laser repeatedly ramping to maximum power at different temperatures for the diode laser and the SHG crystal at the laser head.

Once the pump laser was operational, the SolsTiS-SRX laser was relocated near the pump laser’s head to prevent the divergence of the pump beam from negatively impacting the efficiency of the pump process. A beam pick-off module, provided with the SolsTiS-SRX laser, was then placed close to the pump laser’s output port. This module utilizes a polarizing beam-splitting cube and a half waveplate (see Figure 4.46) to split the pump beam into two paths directed to our titanium sapphire lasers. All components in this module were carefully cleaned using special lens-cleaning tissues and a small amount of ethanol. Cleaning is crucial to prevent residual dirt on the optics, which, given the high-power laser beams, could lead to damage to coatings and potentially cause harm to researchers in the vicinity.

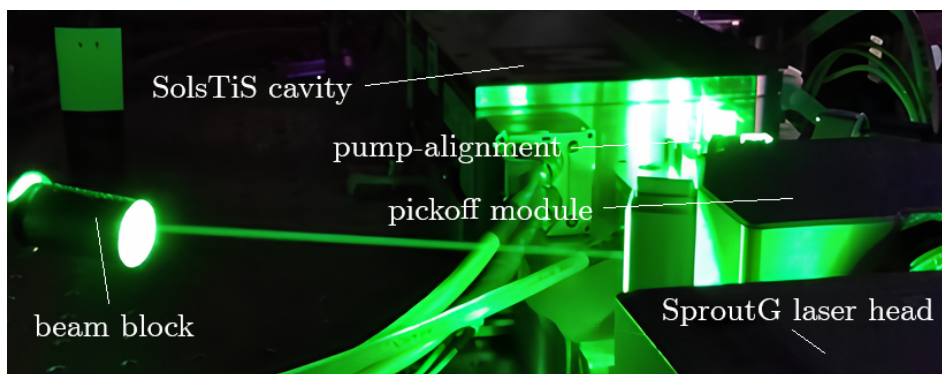


Figure 4.46: Output from the beam pick-off module on installation, where the picked-off beam is blocked for safety measures.

Following the installation of the beam pick-off module, the pump beam alignment module of the SolsTiS-SRX laser was added. This module incorporates two mirrors and a focusing lens. Notably, one of the mirrors within this module is equipped with a motorized mirror alignment mount. When activated, this mechanism automatically optimizes the pump beam orientation to maximize the output power (only when the laser is already lasing). The optical elements in the cavity of this laser are fixed and securely attached to the laser cavity block. Additionally, several

apertures exist within the laser cavity. These apertures eliminate the risk of laser emission in cases of pump beam misalignment. This design ensures the prevention of potential damage to electronics, cables, or optics due to laser emission deviating from the intended optical path.

The pump beam was carefully directed into the cavity by iteratively adjusting the pump beam alignment mirrors until the lasing was observed. The objective during the pump beam alignment was to achieve maximum power through procedural beam walks, ensuring precise alignment with the crystal. After each mirror adjustment, the focusing lens was relocated. Unfortunately, the lens, instead of being mounted on a translational stage, was secured with a set of screws, introducing misalignment due to undesired tilting when the screws were tightened. Consequently, optimizing the output power involved realignment of the lens mounting to minimize undesired tilting. Subsequently, an optimization process for the mirrors was repeated after each lens relocation. This iterative cycle continued until a peak power value was reached, marking the end of the alignment process of the pump beam into the laser cavity. Although the manufacturer's specification sheet indicated an expected output of 4.8 W at 780 nm, we were able to achieve up to 4.3 W, corresponding to 89% of the specified power value. This reduced power output is largely attributed to the deficiencies in the pump beam shape. After passing through the beam pick-off module, it was observed that the pump beam output was distorted. Additionally, the polarization of the beam has elliptical components, which makes the power splitting 92% and 8% when the power is intended to be distributed into one arm only.

SolsTiS-SRX is designed for optimal performance in single-frequency operation, with measures in place to remove the spatial-hole burning effects in the gain medium. The bow-tie ring cavity geometry, coupled with an optical diode, enforces unidirectional operation, which minimizes spatial-hole burning (see Figure 4.47). Wavelength tuning is facilitated by a motorized intracavity birefringent filter (BRF), which introduces wavelength-dependent loss, suppressing wavelengths other than the one the cavity is optimized at. For coarse adjustments, the BRF is employed, while a narrow linewidth operation involves the incorporation of an intracavity thin étalon. This étalon introduces spectral loss, enabling precise frequency tuning by electronically adjusting the étalon spacing. This spectral loss is also selective, further ensuring the suppression of undesired wavelengths. An electronic servo locking of the intracavity étalon provides stability by locking the étalon transmission function to the nearest longitudinal mode. Advanced configurations include a computer-controlled PZT-mounted mirror for precise cavity length adjustments.

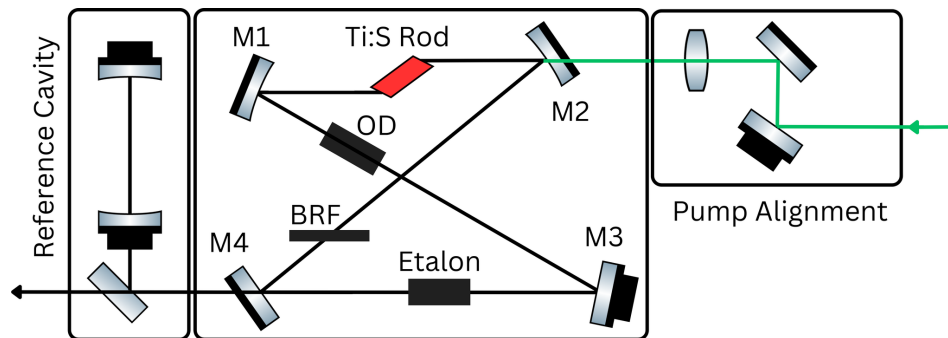


Figure 4.47: Schematic illustration of SolsTiS-SRX intracavity layout and reference cavity for the SolsTiS-SRX. The mirrors with additional thickness represent the PZT mounts. OD: Optical Diode, BRF: Birefringent Filter. *The optical components depicted in this figure are licensed under Ref.[121].*

Further refinement of the laser's linewidth can be achieved by utilization of the external reference cavity of the SolsTiS-SRX laser. In addition, absolute frequency locking to external references, such as atomic absorption lines or optical frequency combs, is achievable. This allows users to fine-tune the output frequency of the laser within the capture range of the locking circuitry. These additional steps ensure the output is not only precise but also stable, providing a high degree of control over the laser frequency.

When tuning the laser frequency to a desired value, users have two options. They can use the GUI, which is an HTML/Javascript-based server running on the laser controller unit. The laser controller unit, when connected via an ethernet interface, provides users with all the necessary operations. Once logged into the server, the GUI allows users to control the birefringent filter angle, etalon voltage, and cavity PZTs. It also enables users to utilize the etalon lock circuit to stabilize the etalon to its nearest longitudinal mode. If an external reference cavity is installed, the laser cavity can be referenced from the GUI as well.

In addition to basic operations, the GUI offers users the opportunity to modify laser electronics settings, such as analog-to-digital converter resolutions or photodiode output amplification, and adjust the parameters of the lock algorithms for stable optical referencing. However, these operations in the GUI are mostly manual, making them less compatible when a sequence of experiments is in progress. For example, in photoassociation spectroscopy of ultracold atoms in a dipole trap, the laser frequency needs to be incrementally adjusted in each iteration of the experimental sequence. These operations can be performed via the '*WebSocket*' communication protocol by sending specific commands to the laser control module [191]. '*WebSocket*' is a bidirectional communication protocol that can send data from the client to the server or vice versa, reusing the established connection channel. This allows tuning the laser cavity PZT at the start of each experimental sequence, ensuring that the laser frequency remains stable when exposing the ultracold sample to the photoassociation light in the dipole trap.

While there are numerous features available in the laser control unit, a particularly useful tool is the ability to link the control unit to a compatible wavemeter. Fortunately, our research group had a compatible wavemeter, '*WS7-60*' of *HighFinesse*, enabling the linkage of the two devices. When linked, the laser controller unit is informed about the output frequency of the light, allowing its tuning algorithm to adjust the light frequency to the desired value. Unfortunately, despite having the opportunity to link the laser control module with the wavemeter, we were unable to operate with this tool due to a malfunctioning PZT element in the external reference cavity.

After the installation, we verified the tuning range of the laser. The wavelength of the laser was systematically adjusted in 5 nm increments, and the corresponding output power was recorded using a power meter. The resulting wavelength-to-output power curve is shown in [Figure 4.48](#). Notably, around 760 nm, there exists a dip attributed to the presence of oxygen and its A-band transition, causing absorption of the laser beam within the cavity and resulting in a decrease in output power [192]. Matching the laser frequency precisely to this transition may cause the laser to shift to a different mode, as the primary mode becomes less dominant than neighboring ones. If necessary, the cavity can be purged with nitrogen gas to eliminate oxygen molecules and reduce the impact on the laser's output power.

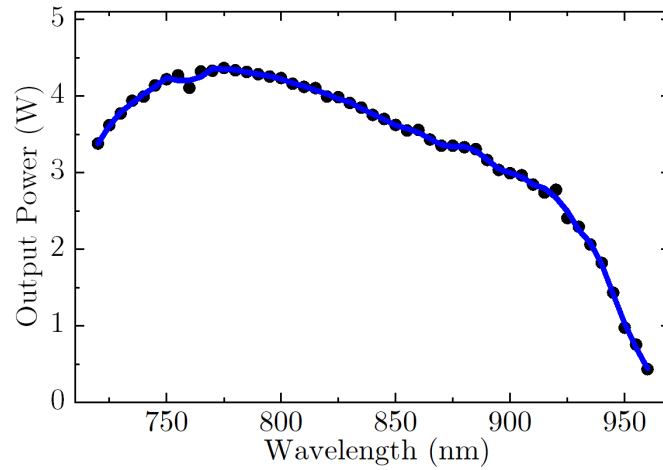


Figure 4.48: Relation between the wavelength and the output power of the installed SolsTiS-SRX laser measured with the pump beam power of 15 W.

The output beam from the SolsTiS-SRX laser is distributed with an optical setup as shown in Figure 4.49. This arrangement divides the beam into two paths immediately after the laser's output port via an achromatic half-waveplate and a polarizing beam-splitter cube. One optical path is directed into an AOM, functioning as a rapid shutter. The output from the AOM is then coupled into an optical fiber to be transmitted to the experimental chamber, where ultracold samples are prepared. An additional optical path is available immediately after the AOM for potential future use.

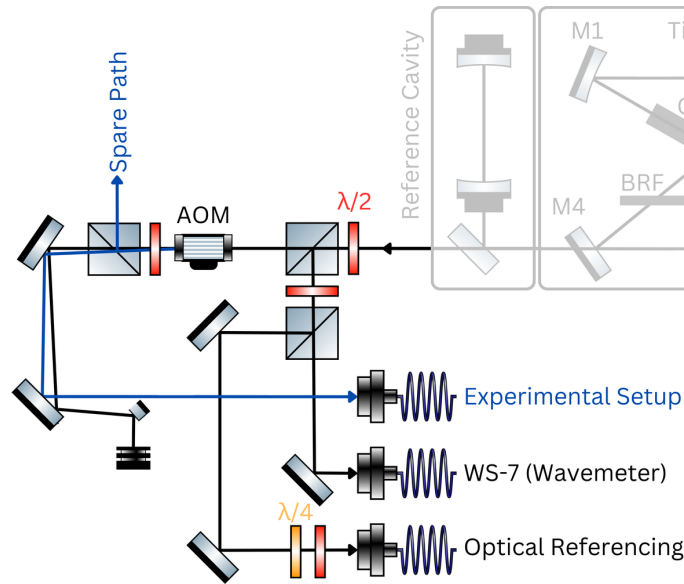


Figure 4.49: Layout of the light distribution setup of the installed SolsTiS-SRX titanium sapphire laser. *The optical components depicted in this figure are licensed under Ref.[121].*

The second optical path undergoes another splitting of the beam, with one path leading to an optical fiber directed to a wavemeter for rough frequency readouts (≤ 60 MHz). The other branch is directed into an optical fiber and sent to another setup designed for optical referencing. Further details about the optical referencing setup are provided later in this section.

The light distribution setup is designed to minimize optical losses on its elements, maximizing the potential of the titanium-sapphire laser. In contrast to the typical double-pass configuration used with tunable lasers, a single-pass configuration for acoustics modulators (AOMs) is employed. While the use of single-pass AOMs becomes problematic with significant changes in light frequency (as the diffraction angle of the beam exiting the AOM is wavelength-dependent), a solution is implemented to address this challenge.

In this setup, the driving frequency of the AOM is tuned to compensate for the change in the diffraction angle when the light frequency undergoes substantial variations. Initially, the AOM is driven with a 110 MHz RF frequency, following the manufacturer's recommended specifications. Subsequently, as the laser output is tuned to 780 nm, the coupling into the fiber is optimized. When tuning the laser to a different wavelength, a decrease in coupling efficiency is expected due to misalignment caused by the wavelength change. To counter this, the AOM's driving frequency is adjusted to achieve maximum coupling efficiency by compensating for the altered diffraction angle. This iterative process is repeated to determine the necessary driving frequencies of the AOM for various wavelengths (see Figure 4.50). Simultaneously, the transmission efficiency of the polarization-maintaining optical fiber (designed for 760-980 nm) is determined. This approach ensures efficient coupling and optimal performance throughout the wavelength range.

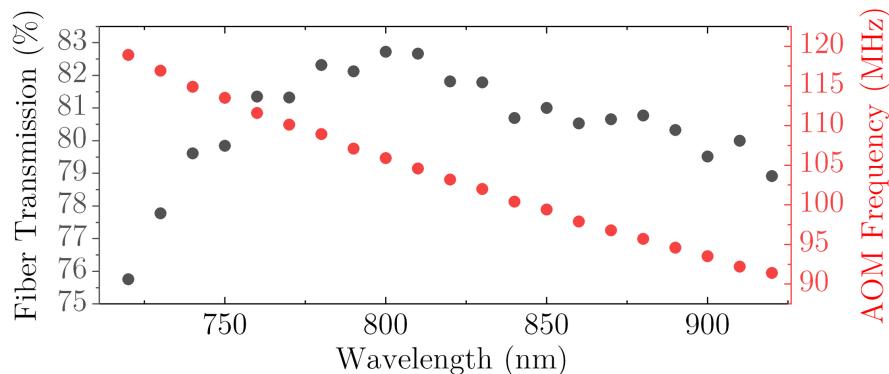


Figure 4.50: A plot showing the relation between the wavelength and the fiber transmission efficiency (left y-axis) and the optimized driving frequencies of the AOM (right y-axis) in the light distribution setup to ensure optimized coupling efficiency into the fiber.

Now that the 'SolsTiS' titanium-sapphire laser is installed and its light distribution setup is detailed, it is worth addressing some issues we encountered and my approach to finding a solution. When the SolsTiS laser became operational, an issue arose when attempting to lock the laser to its reference cavity; the laser frequency could not be successfully locked. To understand the issue, one should know the mechanism behind the working principle of a reference cavity. The locking mechanism involves directing a small fraction of the laser's output to the reference cavity and subsequently locking the laser's output frequency to a reference cavity fringe by tuning the cavity length using a cavity mirror mounted on a PZT.

Upon examination, it was found that the reference cavity had two PZTs; one of them was controlled via the GUI and the other with external analog voltage. Applying external analog signals allowed tuning of the reference cavity, but the laser still failed to lock via the GUI. After checking connections between the laser control unit and the reference cavity, ensuring no connection issues, and tuning the reference cavity PZTs within their working range (0 V to 300 V), it was observed that the GUI-controlled PZT was malfunctioning.

To resolve this, the connections of the two PZTs at the laser control unit were switched. It is highly recommended to determine and label connections beforehand to avoid applying damaging negative voltages to the PZTs. While this made the reference cavity lock via GUI functional, certain features like automatic tuning to the desired frequency, when linked to the wavemeter, remained unresolved due to the different polarities of the two PZTs. This polarity mismatch caused the cavity length to extend when a lower voltage was applied, conflicting with the automatic tuning algorithm's hard-coded design. To address this, we have written a Python script that uses the WebSocket commands and, by doing so, allows real-time wavemeter readouts to be used to tune the laser frequency to the desired values effectively.

With the given details of the '*SolsTiS-SRX*' titanium-sapphire laser and its light distribution setup, we can proceed with the second titanium-sapphire laser '*TIS-SF-777*' manufactured by '*Tekhnoscan Laser Systems*'.

The '*TIS-SF-777*' laser is characterized by a more analog nature when compared to MSquared's SolsTiS-SRX laser, with a relatively simpler control system and fewer digital features in operation. Notably, all of its optics are accessible, removable, and replaceable. Unlike the fixed mounts in the SolsTiS-SRX laser, the mounts of the optical elements in the '*TIS-SF-777*' are not fixed. Additionally, despite having a similar wavelength range, the '*TIS-SF-777*' requires different sets of mirrors and optical elements for three distinct wavelength ranges (650 nm-750 nm, 750 nm-850 nm, 850 nm-950 nm). This presents a significant drawback as replacing optics can lead to misalignments, necessitating a re-installation of cavity elements and potentially resulting in lower laser power outputs. The challenge of storing different sets of optics further complicates the maintenance process. However, it also offers users an opportunity to modify the cavity elements and mirrors if needed. [Figure 4.51](#) illustrates the cavity elements of the TIS-SF-777 and its external reference cavity.

The installation process of the '*TIS-SF-777*' is divided into three main components: Water cooling installation, pump beam alignment, and cavity arrangement.

For the cooling phase, the *NanoTherm3* chiller, previously employed for the SolsTiS-SRX laser, is repurposed to serve both Ti:Sapph lasers. The chiller employs a T-shaped divider to direct water flow into two paths for both input and output channels on the chiller to facilitate logical fluid circulation. Operating at a temperature of 20°C, the cooling system removes heat from both lasers efficiently, with observed temperatures of supply water being consistently around 19.9°C during extended laser operation, confirming the system's reliability.

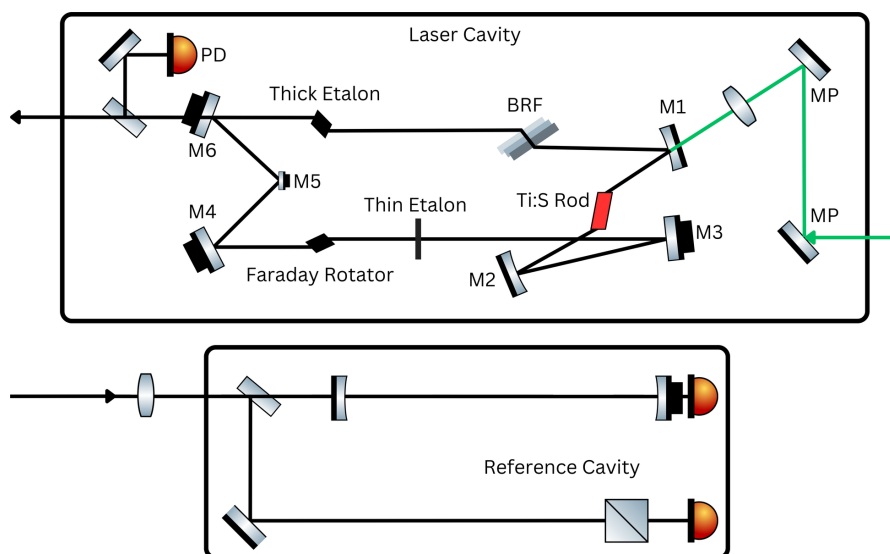


Figure 4.51: Schematic of Tekhnoscan intracavity layout and reference cavity for the TIS-SF-777. The mirrors with additional thickness represent the PZT mounts. PD: Photodiode, BRF: Birefringent Filter. *The optical components depicted in this figure are licensed under Ref. [121].*

The pump beam alignment involves configuring the Sprout-G 15 W laser to support both the SolsTiS-SRX and TekhnoScan lasers. The pump beam is split into two at the SolsTiS-SRX laser's beam-pickoff module, allowing for directed light paths to the SolsTiS-SRX and the TekhnoScan laser, respectively. Tuning the angles of half-waveplates facilitates the selection of the desired path and power arrangement. The TekhnoScan laser's input port, positioned approximately 120 mm above the optical table surface, necessitates careful alignment of six pump beam alignment mirrors in which some of the mirrors are facing upwards. Thus, it is necessary to inspect these mirrors for dust accumulation, considering the laboratory's sub-optimal air quality.

The laser cavity alignment procedure, detailed in the manufacturer's manual, involves iterative steps. Laser cavity elements, excluding mirrors, are first removed, and the laser is configured to operate in linear mode by adjusting mirror mounts. Lasing conditions are achieved through iterative mirror angle alignments, allowing fluorescence spots from the crystal to coincide after being reflected by different cavity mirrors. Subsequently, laser cavity elements are installed one by one, altering the optical path and requiring corresponding mirror angle adjustments. The laser is then reconfigured to operate in ring mode, introducing greater sensitivity to changes in mirror angles. Optimization of output power using cavity mirrors concludes the laser cavity alignment process. Once all cavity elements are installed as in the procedure, it results in a successful installation and alignment of the 'TIS-SF-777' titanium-sapphire laser. The tuning range for the optics set installed for 850 nm to 950 nm is given in Figure 4.52.

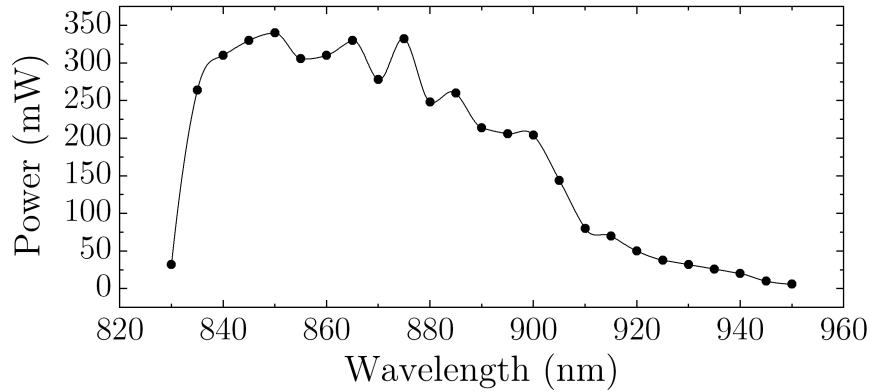


Figure 4.52: Relation between the wavelength and the output power of the installed TIS-SF-777 laser measured with the pump beam power being 15 W.

The ‘TIS-SF-777’ has two electronic control modules. One for laser cavity control is called *Laser Controller Unit*, and the other one is named *Reference Cavity Controller*. Unfortunately, both modules are mostly designed for manual operation, with control knobs placed in the front panel of both electronics modules. In principle, only the Laser Controller Unit is necessary for proper lasing control operation, whereas the reference cavity controller is optional. The ‘TIS-SF-777’ laser has its separate reference cavity module where the output beam from the laser should be aligned to the reference cavity as well (see Figure 4.51).

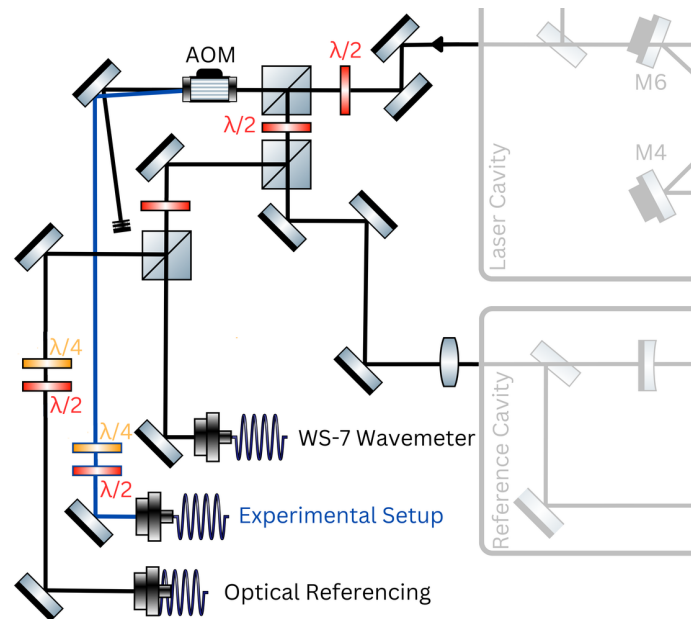


Figure 4.53: Layout of the light distribution setup of the installed ‘TIF-SF-777’ titanium sapphire laser. The optical components depicted in this figure are licensed under Ref. [121].

The connections between the electronics and the laser itself were redone due to the missing cables with specific pin layouts and connection schemes. Due to the absence of a manual or scheme for the connections of the pins, a reverse engineering process was undertaken. There was a modification to the PC7TB Russian standard connector, which was later replaced with a DS-9 Male connector to facilitate standardized usage.

The light distribution arrangement follows the same methodology employed in the SolTiS-SRX laser's distribution setup and is illustrated in [Figure 4.53](#). All output beams are divided and coupled into polarization-maintaining optical fibers. Additionally, a dedicated AOM is incorporated to facilitate control over its intensity, serving as a fast shutter mechanism for the optical pathway directed toward the experimental chamber. The other distinct paths are designated for wavemeter readouts and optical referencing. Furthermore, an optical pathway is directed to the reference cavity, where the laser's synchronization with and stabilization to the reference cavity is achieved.

4.6.2 Referencing to an optical frequency comb

To enhance the accuracy of our laser frequency and, consequently, the photoassociation measurements, we have implemented an optical frequency referencing method for our titanium sapphire lasers. Our HighFinesse WS-7 wavemeter offers an accuracy of 60 MHz or less. To further refine this, the lasers have been referenced to an optical frequency comb. For this purpose, several commercially available PID servos have been implemented to stabilize both the frequency comb and the lasers. The first step involves stabilizing the optical frequency comb itself, followed by optically locking the lasers to this frequency comb.

To initiate our discussion, we briefly state an overview of the optical frequency comb technology. Optical frequency combs have revolutionized precision frequency metrology and enabled advancements in various fields, from spectroscopy to telecommunications [\[193, 194\]](#). At their core, optical frequency combs are comprised of a series of evenly spaced optical frequencies, akin to the teeth of a comb, spanning a broad spectral range. These combs are typically generated using mode-locked lasers, which produce ultrashort optical pulses with a highly stable repetition rate. One of the fundamental concepts behind optical frequency combs is the relationship between the optical frequencies and the time-domain properties of the mode-locked laser pulses. Specifically, the evenly spaced optical frequencies correspond to equally spaced modes in the frequency domain, which in turn arise from the regularly spaced temporal modes of the ultrashort pulses in the time domain. This unique property enables optical frequency combs to serve as a precise frequency ruler or reference, facilitating accurate measurements of optical frequencies and their stability. By referencing lasers to an optical frequency comb, researchers can achieve exceptionally high precision and stability in their experiments, which is crucial for applications such as precision spectroscopy, optical frequency synthesis, and optical clock metrology.

The key properties of an optical frequency comb (OFC) encompass several aspects. Mode-locking initiates the generation of the comb, where all resonant modes of the laser oscillate coherently, producing a series of pulses rather than a continuous beam. The repetition rate (f_{rep}), defining the frequency at which these pulses are emitted, corresponds directly to the spacing between the comb's teeth [\[195\]](#):

$$f_j = f_{\text{ceo}} + j \cdot f_{\text{rep}} . \quad (4.32)$$

With integer values of the index j . This means that there is a so-called equidistant frequency comb, and all occurring optical frequencies are determined by the f_{rep} and f_{ceo} . The carrier-envelope offset frequency (f_{ceo}), representing the disparity between the peak of the pulse envelope and the actual light frequency, crucially influencing the positioning of the comb lines where it can be expressed as [193]:

$$f_{\text{ceo}} = \frac{\Delta\varphi_{\text{ceo}} \bmod 2\pi}{2\pi} f_{\text{rep}} , \quad (4.33)$$

where, $\Delta\varphi_{\text{ceo}}$ is the change in the carrier-envelope offset phase, per resonator round trip, and f_{rep} is the pulse repetition rate. The carrier-envelope offset frequency thus lies between zero and the repetition rate f_{rep} . Ensuring stability in both the repetition rate and the carrier-envelope offset frequency is imperative for the utility of an OFC. This stability is achieved through design considerations and active stabilization techniques, the reliability and precision is essential for further improving the frequency measurements.

In a scenario where both parameters (f_{ceo} and f_{rep}) are precisely known (thus stabilized), the frequencies of every comb tooth become determinable as shown in Equation 4.32. This knowledge allows for the identification of any optical frequency falling within the frequency comb range; for instance, by obtaining a beat note between the OFC and the laser, we want to measure its frequency. The process involves recording a beat note between the unknown frequency and the comb. The lowest beat frequency obtained indicates the distance from the unknown frequency to the nearest comb line. Nevertheless, it fails to provide information about the sign of the detuning from the nearest line, as it could be either above or below the frequency of the comb teeth. The sign of the beat note can be easily determined, for example, by increasing the repetition rate of the comb and observing if this causes the increase or decrease of the beat note frequency. In the former case, the laser's frequency is lower than the frequency of the nearest comb line.

In our case, we had the opportunity to acquire a custom-designed optical frequency comb tailored specifically for the photoassociation of KCs molecules [35]. This optical frequency comb boasts a tunable central frequency range extending from 849 nm to 853 nm. With a spectral width of approximately 7 nm (see Figure 4.54), this custom-made optical frequency comb proves to be optimal for conducting photoassociation spectroscopy of Cs₂ molecules.

The CEO frequency, although not directly observable within the comb optical spectrum, can be detected using a method initially proposed by Telle et al. [196]. This technique employs nonlinear interferometry to generate a heterodyne beat between two harmonic spectral ranges of the comb spectrum [197]. A prerequisite for successful detection is a sufficiently broad comb spectrum so that the harmonic components coincide within the same wavelength range, enabling the detection of the heterodyne beat. One of the most prevalent approaches utilizes f-to-2f interferometry, where a red portion of the comb spectrum is frequency-doubled and then heterodyned with the existing blue comb component. This way, the generated f_{ceo} frequency can be used for the stabilization of the optical spectrum of the comb. In our setup, we feed this generated signal into an electronic circuitry where a digital phase lock loop method is utilized for stabilization as illustrated in Figure 4.55. In our case, the output signal power of this monitoring channel for

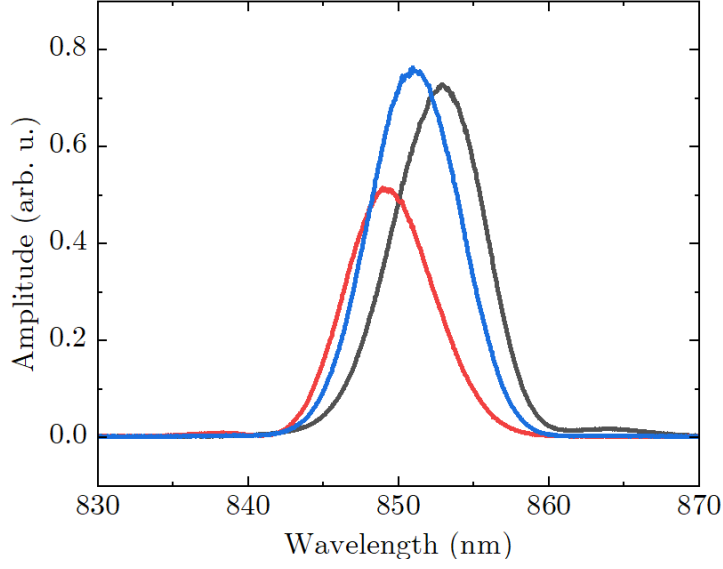


Figure 4.54: Spectrum of the optical frequency comb within its tunability range. Black and red lines represent the spectral profile of the optical frequency comb when at the lower and higher limits of its tunability. The blue line represents the central point of its range.

f_{ceo} is approximately -48 dBm. To enhance the signal, an RF amplifier with a gain of 40 dB is integrated into the circuit. Additionally, a 50 MHz low-pass filter is implemented to eliminate higher harmonics of the obtained signal, where f_{ceo} is typically around 22 MHz in our OFC. This filtering is crucial not only to remove higher harmonics but also to minimize the presence of other frequencies, such as the repetition rate (f_{rep}) and its difference from the CEO frequency, which can introduce unwanted interference and noise into the measurement. The processed signal is then directed to an FPGA. Specifically, the 'RedPitaya' (14-bit version), equipped with firmware designed for phase lock loop operations [198].

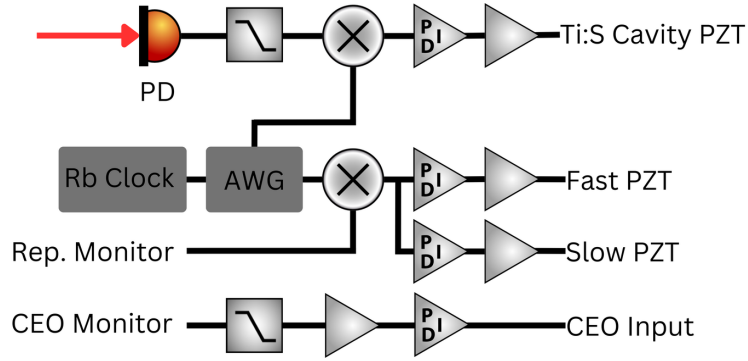


Figure 4.55: Electronic schematic of the stabilization circuitry of our optical frequency comb. The optical and electronic components depicted in this figure are licensed under Ref.[121].

The RedPitaya can operate using its internal clock, but to increase the accuracy of determination of RF signals, we choose to supply it with an external clock. It comes from an arbitrary waveform generator disciplined with an Rb clock (SRS - Rubidium Frequency Std. FS725). The Red Pitaya firmware features a graphical user interface (GUI) developed in Python, which enables full control of the phase-locked loop parameters. Upon launching the GUI, users can establish a connection with the RedPitaya, enabling the device to generate voltage outputs based on provided Proportional-Integral-Derivative (PID) parameters.

The user inputs the desired lock frequency, allowing the system to tune the CEO frequency to a predetermined value. While the FPGA's output is limited to a range of ± 1 V, it proves sufficient for stabilizing the CEO frequency within a narrower range. Although the optical frequency comb accepts control inputs in the range of ± 10 V, amplifying the FPGA output would compromise stabilization quality due to the signal resolution limitations. The stabilization quality of the CEO frequency is determined through a frequency counter, revealing a remarkable precision of 135.48 mHz after 2000 counts of sampling. Additionally, one can see the stabilized spectral profile of the CEO frequency in Figure 4.56. This level of accuracy is particularly notable in the context of utilizing an optical frequency comb for photoassociation spectroscopy. For the photoassociation spectroscopy, as reported in this thesis, the sub-Hertz level stabilization of the f_{ceo} is much better than required. This is because it would introduce only a sub-Hertz uncertainty of the optical frequency determination.

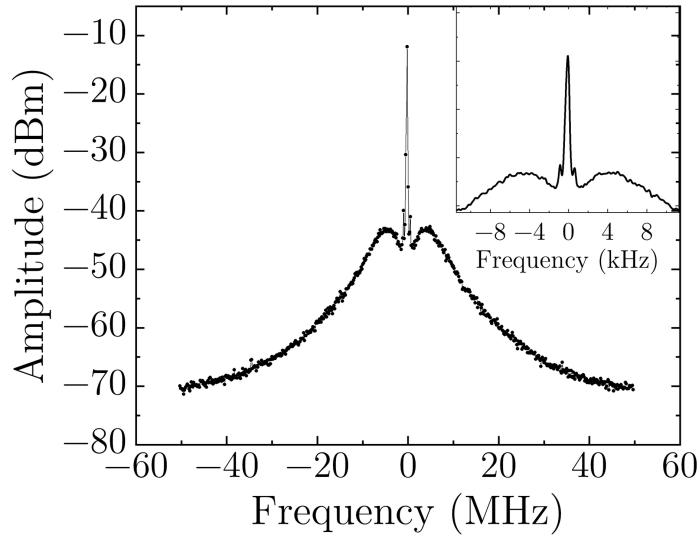


Figure 4.56: Spectrum of the CEO frequency obtained by an RF spectrum analyzer. The x-axis is shifted by the CEO frequency (21.8 MHz) to center the plot on zero.

As emphasized in this thesis, the repetition frequency serves as a defining parameter for the separation of optical frequencies carried in a pulse. Instabilities in the repetition frequency can result in more substantial deviations in the frequencies governed in the optical frequency comb. Considering the millions of optical frequencies present in our optical frequency comb (3.175×10^6 at $\nu = 0$ of $0_g^-(P_{3/2})$), even a slight deviation in the repetition frequency can have significant implications. For instance, a 3 mHz deviation in the repetition frequency can lead to a deviation of 9.525 kHz in the optical frequencies referenced by our photoassociation laser when probing

the lowest lying vibrational level in the external well of the $0_g^-(P_{3/2})$ state. This relationship highlights the remarkable sensitivity of optical frequencies to changes in the repetition frequency, emphasizing the need for precise control and stabilization techniques.

A similar circuitry is employed for the stabilization of the repetition frequency. The repetition frequency of our optical frequency comb operates around 110 MHz, posing challenges when interfacing directly with ‘RedPitaya’ since this specific FPGA does not support RF inputs above 60 MHz. Consequently, the stabilization of the repetition frequency is facilitated with a Phase-Frequency Detector (PFD) supplied by the manufacturer of the optical frequency comb [35]. A PFD, a crucial component in a Phase-Locked Loop (PLL) system, is responsible for comparing the frequency of two input signals and generating an output voltage proportional to their phase and frequency difference. This output serves as an error signal and is utilized in PID controllers.

For the first port of the PFD, an output signal from the AWG, externally clocked by our Rb, is used as the desired repetition frequency. The second port is connected to the monitoring port of the repetition frequency from the optical frequency comb. The output of the PFD is then directed to a ‘LaseLock’ compact laser frequency stabilization electronics module by TEM, functioning as a PID controller with two channels for the slow and fast PZTs of the optical frequency comb. These channels are represented as two PIDs in Figure 4.55.

The output voltage range for ‘LaseLock’ is constrained to ± 10 V while the PZTs used to control the cavity of the frequency comb (for f_{rep} stabilization) accept voltages in the range 0 V-250 V. Consequently, an analog voltage amplifier for PZTs (‘miniPiA 103’) is utilized. This specific amplifier has gain and offset control knobs with three inputs and outputs, matching the necessary number of amplifiers employed in our optical referencing circuitry for controlling all the PZTs in the setup.

The gain and offset values are set through the following procedure. Initially, PID outputs are manually selected to send 0 V to the *miniPiA 103* amplifier. The offset knobs are then tuned for each channel to meet the mid-point of the range of the PZTs’ working voltage. For instance, the slow and fast PZTs have a working range of 0 V to 250 V; thus, the offset value is set to 125 V. Subsequently, the PID outputs are set to a maximum value of 10 V. By doing so, the gain of the *miniPiA* channels is tuned to supply 250 V at the output. For verification, the PID controller is set to give -10 V, and the corresponding voltage at the amplifier’s output is observed to supply 0 V.

Once the amplification and offset values are set, PID parameters driving the slow PZT are tuned. For each change made in the PID settings, the standard deviation of the repetition frequency is analyzed. By iteratively tuning the PID settings, the optimum PID parameters are found. However, the slow PZT alone is insufficient for stabilizing the repetition frequency at a desirable level, necessitating the utilization of the fast PZT.

When both PZTs are driven simultaneously, interference between them significantly reduces the quality of stabilization. Fortunately, the ‘LaseLock’ module allows users to set a threshold value for PIDs to be activated. When this threshold value is reached in the error signal, the PID output becomes available. Below this threshold, the fast PZT is solely responsible for repetition frequency stabilization, discarding the possibility of interfering tasks for both PIDs. Moreover, if the error signal exceeds the threshold value, the slow PZT is activated. However, as the slow PZT no longer actively contributes to the stabilization process, except for facilitating the oper-

ation of the fast PZT within its working range, its sole responsibility becomes initiating its own deactivation. Moreover, it must do so with a sufficient margin to prevent getting stuck in a phase where the error signal fluctuates around the threshold value. To achieve this, the proportional parameter of its PID settings was intentionally over-tuned. Consequently, the slow PZT was assigned the task of intentionally overshooting its stabilization whenever the threshold value was reached by the error signal. Thus allowing to have a safe margin for avoiding the error signal to fluctuate around the threshold value. When the optimum settings are determined, the stability of the repetition frequency was determined to have a standard deviation of 2.73 mHz, which translates into ~ 8.19 kHz of frequency deviation for a given comb tooth around the investigated photoassociation spectra in this thesis. The spectral profile of the repetition frequency is given in [Figure 4.57](#)

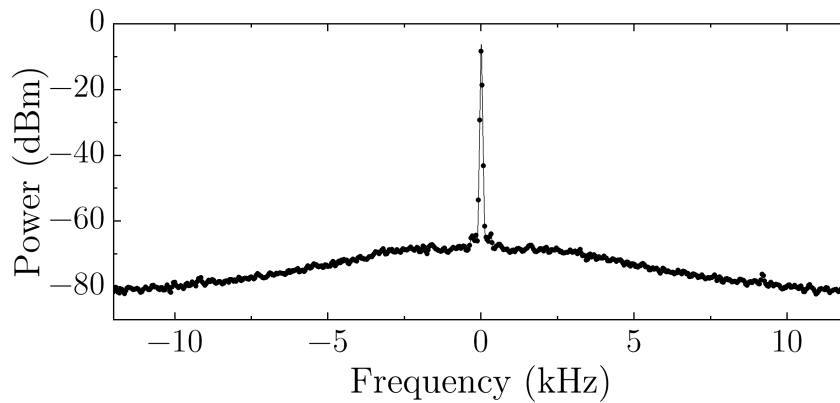


Figure 4.57: Spectrum of the repetition frequency obtained by an RF spectrum analyzer. The actual frequency of the repetition frequency is centered around 110.117032 MHz.

Upon achieving stabilization of the frequency comb, the photoassociation lasers (SolsTiS-SRX and TIS-SF-777) were optically referenced to the frequency comb via stabilization of the optical beat note between the laser and the nearest tooth of the frequency comb. The schematic layout used to generate the desired beat note is illustrated in [Figure 4.58](#). To ensure long-term stability, the beat note signal was extracted using a photodiode connected to an optical fiber, guaranteeing spatial overlap of the laser beams on the photodiode.

The photodiode signal was subsequently amplified by an RF amplifier and fed into another ‘RedPitaya’ (14-bit version), whose clock source was synchronized with our Rb clock. The PID settings of the FPGA were tuned for optimal performance. The lock frequency was set at 30 MHz, resulting in the stabilization of the beat note signal at 30 MHz. Both SolsTiS-SRX and TIS-SF-777 lasers have external control voltages for their PZTs ranging from ± 4 V. The RedPitaya’s output was further amplified with the miniPiA amplifier to accommodate the voltage range. While both lasers feature slow and fast PZTs, using only the slow PZTs yielded a laser linewidth below 50 kHz for SolsTiS-SRX and around 130 kHz for TIS-SF-777. The standard deviation of their central peaks was approximately 200 Hz.

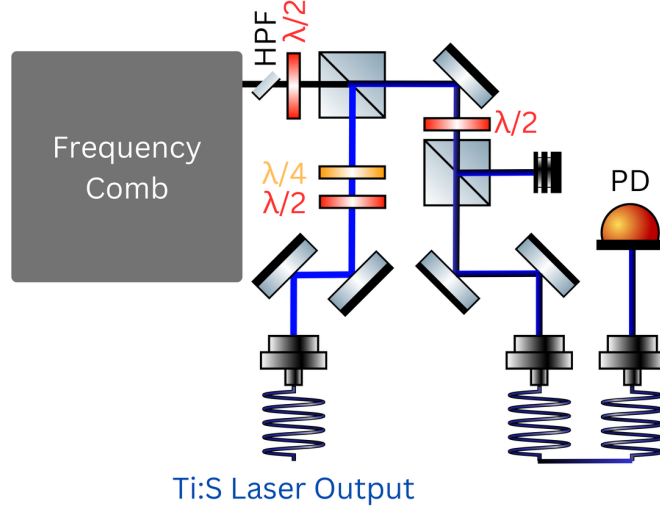


Figure 4.58: Layout of the beat note setup for referencing photoassociation lasers. HPF: high-pass filter, PD: photodiode. *The optical components depicted in this figure are licensed under Ref. [121].*

The frequencies of the titanium-sapphire lasers are tuned by varying the repetition rate frequency of the optical frequency comb. In each iteration of an experimental sequence, Equation 4.34 and Equation 4.35 are employed to determine the frequency of the referenced lasers. To do so, we utilize a combination of measurements and calculations. Initially, we start with determining the referenced comb tooth by the expression:

$$j = \frac{f_{\text{wm}} - f_{\text{ceo}} - f_{\text{lock}}}{f_{\text{rep}}} \text{ where } j \in \mathbb{Z}^+, \quad (4.34)$$

where, f_{wm} represents the wavemeter reading, f_{lock} denotes the locking frequency. By substituting these values into the equation and solving for j , an integer representing the corresponding comb tooth number is obtained. The calculated j number is rounded to the closest integer as j is a positive integer. Once the j comb tooth number is obtained, the frequency of the laser beam sent to ultracold atoms is determined:

$$f_{\text{laser}} = j \cdot f_{\text{rep}} + f_{\text{ceo}} + f_{\text{lock}} + f_{\text{aom}}. \quad (4.35)$$

It is crucial to acknowledge the impact of AOMs on the laser's frequency for the preparation of photoassociation beams. To incorporate this shift, we include f_{aom} in Equation 4.35, representing the frequency alteration introduced by the AOM. This ensures that the frequency sent to the atoms is accurately determined, accounting for all relevant factors.

Chapter 5

Photoassociation of Cs₂ molecules

In this chapter, the systematic progression is detailed, starting from the initial observation of previously probed photoassociation lines of Cs₂ within the magneto-optical trap to the execution of precise spectroscopic measurements aimed at uncovering previously unobserved photoassociation lines in an optical lattice.

Initially, after our research group achieved an operating MOT for cesium atoms and potassium isotopes (³⁹K, ⁴⁰K, ⁴¹K), a project aiming to create KCs molecules with photoassociation spectroscopy was initiated. As described and detailed in [Chapter 4](#), we have made operational two titanium-sapphire lasers to be used in photoassociation experiments, and progressively, we have improved our sample preparation stage for cesium atoms facilitating spin-polarization process, observing Feshbach resonances and also improving the stability of the experimental setup. In preparation for investigating KCs molecules in excited states, we have focused on benchmarking our setup by conducting photoassociation spectroscopy measurements on ultracold cesium atoms. This seemingly routine yet essential initial step, however, yielded unexpectedly fruitful results and led us to uncover new scientific insights with cesium dimers that we had not anticipated.

In our efforts on these benchmarking experiments, we primarily investigate the external well of the $0_g^-(P_{3/2})$ state of Cs₂, which exhibits a double minimum potential. This state has been used to create the first stable ultra-cold molecules [117], and it has appropriate properties to be used in a scheme as it appears as a suitable intermediate step for the formation of the triplet ground state molecules in their $\nu = 0$ vibrational level [23, 102]. Previously, the $0_g^-(P_{3/2})$ state of Cs₂ has been examined by various research groups [20, 25, 21, 22, 24]. Inconsistencies in the parameters obtained from the analysis of the previously obtained experimental data were subsequently addressed by a theoretical work of Bouloufa et al. [23]. The reexamination of this potential led to the conclusion that it should host two additional vibrational levels below what had been up to then considered to be the ground level of this external well. Substantial efforts have been made to observe and identify these ‘missing’ vibrational levels, aiming to resolve inconsistencies arising from earlier experiments [24, 23]. The pursuit of measuring these levels seemingly concluded with the published experimental work of Zhang et al. [24]. Afterward, a reevaluation of these vibrational levels was conducted [25], revealing experimental data that underscored a significant difference in the energy values from the initially reported values of the missing levels. Surprisingly, there has been no commentary to date addressing the disparities

between the two sets of measurements. Moreover, the lowest vibrational levels reported in the Ref. [25] and [24] were observed at energies where the vibrational levels of the $0_u^+(P_{3/2})$ state were also reported to be present. These unexpectedly coincidental results have given us the push to verify the previous measurements reported in Refs. [24, 25].

This chapter begins by detailing the measurements of Cs_2 photoassociation in a magneto-optical trap (Section 5.1). Employing photoassociation spectroscopy within a MOT is deemed a preferred initial step when investigating new molecules. This preference stems from the simplicity of measurements within a MOT, which eliminates the complexities introduced by additional cooling methods. Consequently, obtaining the spectroscopy data is rather fast since only systematically varying the frequency of the photoassociation laser is sufficient as the MOT is constantly being loaded from hot background gas.

Moving forward, in Section 5.2, information on the photoassociation process in an optical dipole trap for Cs_2 is presented. The transition from the MOT to the optical dipole trap introduces higher densities of ultracold samples, thereby establishing a superior environment for conducting spectroscopic measurements. Detection of photoassociation events is achieved by monitoring atom loss from the trap, rendering the measurements particularly sensitive to the formation of excited molecules.

Furthermore, optical dipole traps offer the ability to apply external magnetic fields to manipulate collisions between atoms. Additionally, the optical confinement allows us to prepare spin-polarized atomic ensembles, as discussed in Section 5.3. A crucial part of this thesis work involves the utilization of Feshbach-optimized photoassociation, covered in Section 5.4, where Feshbach resonances are exploited to enhance molecular association rate [135], enabling observation of photoassociation spectra that would otherwise remain below the noise level.

In Section 5.5 and Section 5.6, we present our results on photoassociation spectroscopy and analysis of the molecular states under study. At the same time, we provide clarification regarding the lowest vibrational levels of the $0_g^-(P_{3/2})$ state and the neighboring vibrational levels of the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. Our measurements reveal that the two vibrational levels previously attributed to the $0_g^-(P_{3/2})$ state ($\nu=0$ and $\nu=1$) by Zhang et al. [24] and Ma et al. [25] actually correspond to the $0_u^+(P_{3/2})$ state, contrary to prior assumptions. The analysis of the results we obtained can be found in these corresponding sections.

It should be noted that in this work we use the vibrational numbering of the electronic states $0_g^-(P_{3/2})$, $0_u^+(P_{3/2})$, and $1_g(P_{3/2})$ as follows. For the $0_g^-(P_{3/2})$ state, vibrational numbering in the external well of the $0_g^-(P_{3/2})$ state is done independently from the inner well, in which $\nu = 0$ represents the lowest vibrational level in the external well. In this thesis, the observation of two additional vibrational energy levels at the bottom of the potential well prompts a revision in the vibrational numbering for the $0_g^-(P_{3/2})$ state. The new numbering scheme is given by $\nu_{\text{new}} = \nu_{\text{old}} + 2$. Conversely, for the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states, $\nu' = 0$ corresponds to the highest vibrational level. Their vibrational levels are labeled with $\nu' = [\nu_D - \nu]$, where ν_D denotes the ‘true’ vibrational quantum number of the last vibrational level closest to the dissociation limit. These notations are employed for improved readability, aligning with traditional numbering conventions as found in Refs. [23, 25].

5.1 Photoassociation in a magneto-optical-trap

The photoassociation spectroscopy of ultracold atoms in a MOT has been a very powerful tool to study cesium dimers in electronically excited states [22, 24, 25, 36, 43, 199]. When it comes to the design of our MOT, we cool atoms from the hot background gas available within the same experimental chamber as our MOT. Thus, our MOT is constantly loaded from the hot background gas of cesium atoms. In the context of photoassociation, a photon with energy that matches the energy difference between the pair of two atoms and an excited state molecule forms a weakly bound molecule in the excited electronic molecular state. This process for the photoassociation of Cs_2 to a state converging to the $(6S_{1/2} + 6P_{3/2})$ limit can be shown as:

$$2 \cdot \text{Cs}(6S_{1/2}, F = 4) + h(\omega_0 - \Delta_L) \rightarrow \text{Cs}_2[\Omega_{g,u}, \nu, J, (6P_{3/2})]. \quad (5.1)$$

In which the ω_0 is the frequency of the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$, the Δ_L is the detuning of the photoassociation laser from this transition and $\Omega_{g,u}$ is the labeling of various electronic states of the molecule, as described in [Section 3.2.1](#) and [Section 3.2.2](#).

Since the trapping conditions are fulfilled in our MOT only for cesium atoms but not for molecules, every time a molecule is formed, two atoms are lost from the trap. We monitor the population of atoms by recording the fluorescence they emit during the cooling process. The emitted light is detected via a photodiode within our imaging setup. When molecular formation occurs, the resulting fluorescence signal from the cesium atoms decays because once a molecule is formed, it is not excited by the cooling laser. We can describe the number of atoms present in the MOT by the following expression [200]:

$$\frac{dN}{dt} = L - \gamma N - 2 \int G(r) n^2(r, t) d^3r. \quad (5.2)$$

In this context, L denotes the rate at which particles are loaded into the trap, γ represents the rate of one-body losses unrelated to the photoassociation process, and $G(r)$ signifies the loss rate coefficient arising from photoassociation of the atoms confined within the trap, thus, the last term represents the loss of two atoms per collision event. The density of atoms confined within the MOT can be quite well approximated by the Gaussian distribution $n(r, t) = n_0(t)e^{-r^2/w^2}$, where $n_0(t)$ is the time-dependent peak density, and w is the width of the sample. The function $G(r)$ can be characterized as $G(r) = G_0e^{-\rho^2/\omega^2}$, where ρ denotes the distance from the propagation axis of the photoassociation beam.

In a real-life scenario, the parameters mentioned above play a crucial role in determining the photoassociation rate of a molecule to a targeted vibrational level. However, the photoassociation rate in this model is calculated when the number of trapped atoms in the MOT reaches its equilibrium state. Thus, the frequency of the photoassociation laser should be varied slowly enough to let the MOT reach its equilibrium state whenever a photoassociation process is present.

Moving forward, the quality of the photoassociation spectra depends highly on the loading rate, the density of the atomic ensemble, and how well the photoassociation laser beam overlaps with trapped atoms in the MOT. From an experimental perspective, most parameters involved in

Equation 5.2 are linked to each other. For instance, one can lower the loading rate of atoms into the trap. Doing so, for example by decreasing the current of the getter (atom dispenser), the total number of atoms present in the MOT decreases significantly. This also reduces the cloud size and the number of atoms present in the trap, which reduces the signal-to-noise ratio of readouts and the overlap of the photoassociation beam on the sample. Another example can be given as follows: We can compress the MOT by increasing its magnetic gradient, which is achieved by simply increasing the current flowing through the coils used for generating the quadrupole magnetic field. In such a scenario, the cloud will become denser for a short period since all the cold atoms will be quickly drawn to the center of the trap. However, this usually causes increased losses, and the number of atoms quickly drops to reach a new equilibrium level. This constraint not only reduces the photoassociation rate but also hampers the detection of the fluorescence signal, bringing our readouts closer to the noise level as in the previous example. So far, I have given examples of the variables that can be easily changed via our experimental control system. However, there also exist such variables that are rather unfeasible to tune or time-consuming to vary in general. An example can be the photoassociation laser intensity, where the power has its technical upper limit, and the beam size cannot be easily changed without time-consuming realignment of the optical path. In such experimental conditions, finding an optimized condition is rather tricky. In Equation 5.2, the variable $G(r)$ also depends on the width of the laser beam, which ideally should be large enough to overlap with the cloud size of ultracold atoms trapped in the MOT. Thus, it will eventually end up sacrificing intensity by enlarging the beam or reducing the cloud size by sacrificing the number of atoms, thus possibly decreasing the signal-to-noise ratio. In the case where the beam size is reduced to gain intensity, then the photoassociation process only occurs in a small portion of the cloud, which reduces the observability of many photoassociation lines since, yet again, the loss rate due to photoassociation becomes too low to be identified within the dominant fluorescence signal extracted from the atoms which do not participate in the photoassociation process.

For the optimization of the photoassociation readouts, I have initially targeted a previously known vibrational level, namely, $\nu = 58$ in the external well of the $0_g^-(P_{3/2})$ state. This particular level was chosen since the detuning of this level from the atomic transition is relatively low, thus yielding a high molecular formation rate, and it was previously studied in the reference work of Ref. [201]. Besides in the past, the external well of the molecular potential of $0_g^-(P_{3/2})$ state was examined extensively by several other groups [21, 23, 42, 186, 201, 202] giving us great guidance on improving our experimental apparatus and understanding the behavior of the photoassociation process.

In these experiments, about 20×10^6 cesium atoms were trapped in the MOT, with the majority predominantly occupying the $F = 4$ ground state level. The temperature of the trapped atoms was around $100 \mu\text{K}$. The initial step was to align the photoassociation laser through atoms trapped in the MOT. The alignment process involved reducing the laser power to less than 1 mW and tuning the laser frequency to a resonant frequency of the D_2 line, $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$. The power was reduced via the AOM installed in the distribution setup introduced in Section 4.6. Due to the exposure to resonant light, the cooling conditions were disturbed, and the atoms were depleted from the trap. By lowering the power and adjusting the direction of the laser beam, the alignment process was carried out. For further precision, I iterated this process with the $6s^2S_{1/2}(F = 3) \rightarrow 6p^2P_{3/2}(F' = 4)$ transition, which yielded a weaker response of the cloud thus, beneficial for achieving better alignment. To ensure precise focusing of light in the center of the cloud, I tuned the laser frequency to closely match the transition frequency between the $\nu = 58$ vibrational level of the $0_g^-(P_{3/2})$ state and the unbound

pair of atoms. In the first attempt of detecting a photoassociation signal and consequently being successful at generating Cs_2 molecules in our trap, I have fixed the frequency of the photoassociation laser to target the $J = 2$ rotational energy level of this vibrational level. The decision to prioritize the third rotational level ($J = 2$) was grounded in the observations from previous experiments [22, 203, 204, 205, 206], consistently demonstrating higher effectiveness in facilitating photoassociation across all vibrational levels in the $0_g^-(P_{3/2})$. After making fine adjustments to the direction of the laser beam, I finalized the alignment of the photoassociation laser beam by optimizing the resulting photoassociation rate.

By optimizing the parameters such as the magnetic field gradient, temperature of the cesium getter, and photoassociation beam intensity, I have identified optimum trapping conditions for our readouts of photoassociation spectroscopy data. It should be noted that the optimum parameters depend on the vibrational level of the associated molecule. For the $\nu = 58$ vibrational level in the $0_g^-(P_{3/2})$ state, the spectral data was obtained when the laser power was kept around 300 mW with the beam diameter being 400 μm . However, for the detection of weaker lines, the power was increased up to 2 W. The last observable photoassociation line in the $0_g^-(P_{3/2})$ state in experiments carried in the MOT terminated on vibrational level $\nu = 12$, which is located at the frequency detuned by -60.74 cm^{-1} from the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$. The remaining photoassociation lines below the $\nu = 12$ remained undetected. The external well of the $0_g^-(P_{3/2})$ state of Cs_2 has numerous documented experimental data from studies conducted in both the MOT and the optical dipole trap. Considering the significant time investment required for extracting spectral data across hundreds of vibrational energy levels, measurements in the MOT have been kept to a minimum.

In Figure 5.1, the photoassociation spectrum corresponding to excitation of vibrational level $\nu = 58$ in the $0_g^-(P_{3/2})$ state is presented, showcasing well-resolved rotational lines. The photoassociation experiments in the MOT followed a straightforward procedure. Upon aligning the photoassociation beam, the MOT was loaded to maintain a steady-state number of atoms. Subsequently, the frequency of the photoassociation beam was adjusted to the region of interest in the spectrum, and then the beam was illuminated on the sample.

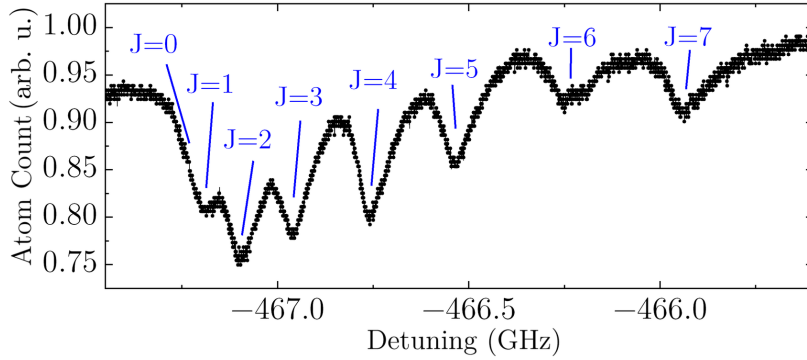


Figure 5.1: Trap loss spectrum of the photoassociation line corresponding to $\nu = 58$ of the $0_g^-(P_{3/2})$ state. The frequency detuning is with respect to the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$.

Readouts obtained from the photodiode were synchronized with the output of the wavemeter

‘WS-7-60’ manufactured by ‘High Finesse’. The wavemeter was configured to send analog voltage signals calibrated to a reference frequency point, treating this as 0 V. Depending on the user-set gain value, the wavemeter supplied corresponding voltages in real time, synchronized with the readouts. Combining the analog output from the wavemeter with the fluorescence signal readouts from the photodiode allowed for the synchronization of the obtained photoassociation spectra with the laser frequency. Figure 5.2 illustrates this synchronization, displaying the photodiode signal presenting the photoassociation spectra alongside a straight line indicating the frequency of the photoassociation laser. The recorded data facilitated the translation of the voltage output from the wavemeter into frequency, which was then integrated with the data obtained from the photodiode readouts.

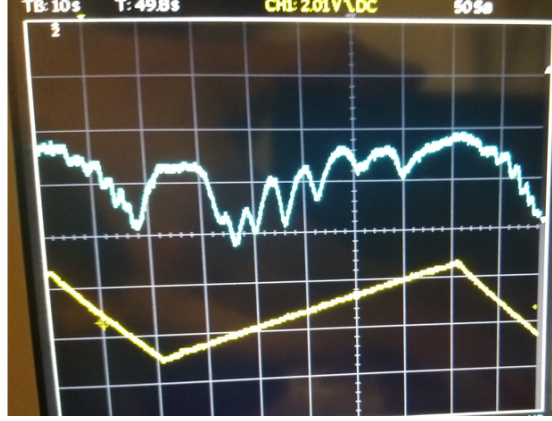


Figure 5.2: A snapshot of the oscilloscope captures the recording of fluorescence signals, representing the trap-loss-based photoassociation spectra (cyan data points). Simultaneously, it displays the analog output from the wavemeter readouts, depicted as yellow data points, providing a direct translation into the frequency of the photoassociation laser.

The rovibrational spectra of the measured vibrational levels give us additional clues through the line strengths of the rotational lines. The allowed values of rotational numbers are $J = (l - 2), l, (l + 2)$, where l characterizes a partial wave of the continuum state and $\vec{J}$ is the sum of $\vec{l}$ and of the total angular momentum of the electrons [34, 207]. In the spectra shown in Figure 5.1, one can see that due to the angular momentum conservation of pure s -wave scattering, the $J = 2$ becomes the strongest line [13]. The s -wave scattering, in fact, can be observed even when the sample temperature is close to 0 K, while the other partial waves do not play a role. The appearance of higher rotational levels can be explained by higher angular momentum contributions, such as orbital angular momenta arising from p -wave, d -wave, and so on. Given the fact that our trapped cesium atoms in the MOT have an average temperature of 100 μ K, observation of the effects of higher partial waves is expected.

5.2 Photoassociation in an optical dipole trap

The utilization of optical dipole traps provides unique capabilities for ultracold experiments. Not only do they offer higher sample densities, but they also enable the application of external magnetic fields to trapped atoms, crucial for tuning their collisional properties. Given the availability

of our optical dipole trap for photoassociation experiments, we have not pursued the search for vibrational levels $\nu < 12$ in the $0_g^-(P_{3/2})$ state using the MOT.

Confining ultracold atoms in an optical dipole trap makes the experimental setup more complex. Detailed information on the cesium cooling lasers and dipole trapping laser is provided in [Chapter 4](#). Below, I present an overview of the photoassociation measurements used in our optical dipole trap setup.

Photoassociation measurements were conducted using a 1D optical dipole trap created by a 50 W red-detuned 1064 nm laser (*NuAmp 50 W* manufactured by *Nufern*). The cesium sample, prepared with the MOT, was cooled down to 10-12 μK using gray-molasses cooling (GMC). Subsequently, the sample was loaded into the optical dipole trap formed by crossing two beams with a diameter of 250 μm at an angle of 140° . The trapped atoms had a cylindrically symmetric Gaussian density distribution, with typical dimensions of 150 μm radially and 600 μm axially. The lifetime of cesium atoms in the dipole trap was approximately 5 seconds, primarily influenced by collisions with the hot background gas. While this poses a challenge for experiments requiring longer duration (in particular if evaporative cooling is involved), it is typically sufficient for photoassociation experiments, which usually take around 1 to 2 seconds.

For photoassociation, we employed a commercially available titanium:sapphire laser (*MSquared* - *SolsTiS-SRX*) with a maximum output power of 3.8 W around 852 nm and a linewidth of $\Delta\nu \leq 50$ kHz. An AOM was incorporated to facilitate rapid control of the power and illumination time of the cesium sample. A portion of the laser beam was directed to a wavemeter via an optical fiber for frequency readout with an absolute accuracy of less than 60 MHz.

Trap-loss-based fluorescence readouts were employed to acquire photoassociation spectra in our study. This involved the controlled release of atoms from the dipole trap, the subsequent application of optical molasses, and the collection of fluorescence data. Our methodology prioritized the observation of changes in fluorescence levels as a proxy for variations in atom number due to trap loss, eliminating the need for precise atom quantification. The observability of trap-loss spectra depends primarily on trap depletion dynamics [13, 208], emphasizing the importance of consistent temperature and repeatable atom number in each iteration of the experimental sequence. Compared to [Equation 5.2](#), the trap loss model here can be described in a simpler way with no loading rate being present in the dipole trap:

$$\frac{dN}{dt} = -\gamma N - 2 \int G(x) n^2(x, t) d^3x, \quad (5.3)$$

where, γ represents the one-body loss rate unrelated to the photoassociation process and $G(x)$ signifies the loss rate coefficient arising from the photoassociation of atoms confined by the trap. The model originates from the work of S. Kraft et al. [13, 20] and is explained in detail there. In brief, the proposed photoassociation process model suggests that the loss of atoms in the optical dipole trap is highly influenced by the intensity of the photoassociation laser beam and the density of the sample. From here, one can determine the photoassociation rate coefficient of a specific rotational line.

The trap-loss model described above also depends on the intensity profile of the photoassociation beam passing through the ultracold sample. In the initial stages of our experiments, the photoassociation beam was directed through the center of the sample along its longer side. However, this

configuration presented alignment issues and challenges due to the size of the photoassociation beam (with a diameter of approximately $200\text{ }\mu\text{m}$). The beam overlapped with only one-third of the axial length of the atom cloud, causing the photoassociation process to occur only in this limited region (see Figure 5.3). This limitation diminished the quality of the obtained photoassociation spectra.

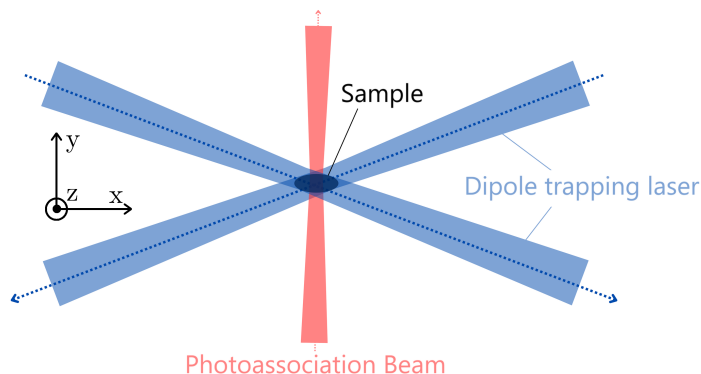


Figure 5.3: An illustration of the optical paths for optical dipole trapping beams (blue) and the photoassociation laser beam path (red). The photoassociation beam here is in the ZY plane, at about 45° to the Z axis.

In addition to the aforementioned challenges, we have detected distortions in the spatial beam profile of the dipole-trapping laser. This led to the formation of local intensity maxima at various positions within the focal region, consequently diminishing atom density and compromising the quality of photoassociation spectra by creating localized regions of atom accumulation. In the later phases of this thesis, we transitioned to employing the ‘*Azurelight 50 W*’ amplifier for our dipole trapping laser, which exhibited a spatial profile closely aligned with the specifications outlined in the product documentation.

Considering these negatively influencing conditions, we first attempted to measure photoassociation lines with the assessment of the remaining relative atom number through trap-loss-based measurements, as depicted in Figure 5.4a. However, the obtained spectra lacked resolution, prompting us to explore alternative methodologies. Subsequently, we investigated the temperature dynamics of the sample by monitoring the cloud size after a 2 ms expansion period, as illustrated in Figure 5.5b. This expansion allowed us to observe the heating effect induced by the photoassociation process. The rationale behind this approach stems from the possibility that some molecules may dissociate into free atoms, which still remain trapped. Although not reflected in trap-loss measurements, these atoms possess higher kinetic energy, contributing to the heating of the sample. Such heating can arise from collisions between associated molecules and cold atoms or between trapped atoms and high-energy atoms emerging from dissociated molecules. This scenario is not uncommon, especially when dissociation serves as the primary decay channel for formed molecules within a deep confining potential.

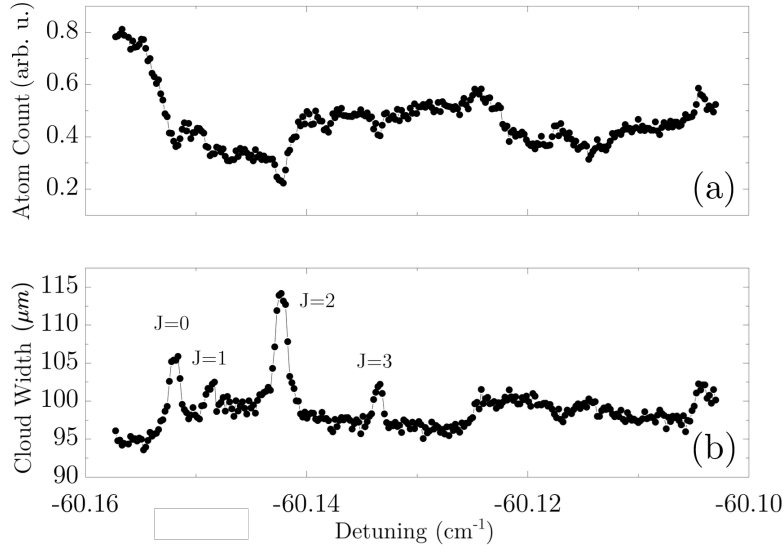


Figure 5.4: a) Trap loss spectra of the vibrational level $\nu = 12$ versus the detuning from the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$. Each data point is obtained after the completion of a full experimental sequence with a total of 270 experiments. b) The size of the cloud is determined at 2 ms after being released from the optical dipole trap versus the detuning from the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$.

Early experiments revealed large fluctuations in the atom number trapped in the dipole trap. The cause of these fluctuations was investigated for a while, but the source was not identified for an extended period of time. Finally, we decided to track the long-term stability of the cesium cooling lasers by monitoring the power of MOT beams close to the experimental chamber. As such, our measurement became sensitive to possible polarization drifts. They could occur because each cooling beam (cooling and repumper) is delivered to the experimental table via a single-mode, polarization maintaining optical fiber (see [Section 4.2.1](#)), and then the beams were split with polarizing beam splitting cubes into different optical paths required for various cooling stages. Therefore, any observed power drift in this scenario would have to come from polarization fluctuations, as we already knew that the power coming out of the fibers did not fluctuate. With this information in hand, we identified that the polarization of the lights that were coupled into the cooling and the repumping fibers was slightly elliptical. As a result, the light coupled into the polarization-maintaining fiber propagated along both the slow and the fast axis, experiencing different phase shifts that depended on the environmental conditions (temperature, mechanical vibrations, humidity). It manifested itself as polarization fluctuations at the fiber output. After adding quarter-waveplates to make the polarization before the corresponding polarization-maintaining optical fibers linear, the fluctuations were eliminated.

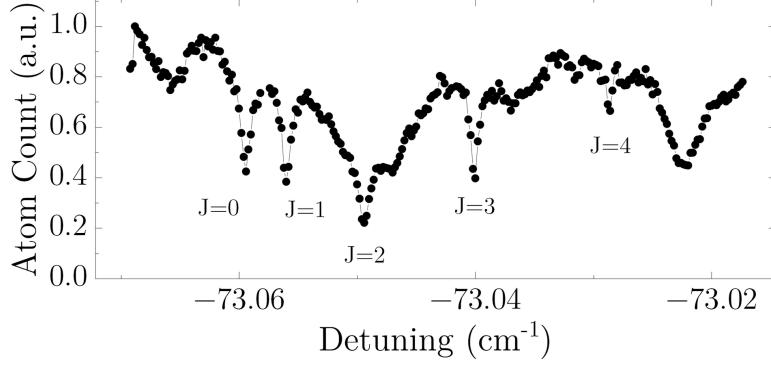


Figure 5.5: Trap loss spectra of the vibrational level $\nu = 4$ of the $0_g^-(P_{3/2})$. Its rotational levels $J = 0$ to $J = 4$ are well resolved. The detuning is given in reference to the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$.

A significant improvement was achieved with the implementation of the lock-in detection scheme combined with a variable timebase in our setup. This implementation enabled us to create experimental sequences that incorporate conditional loading of the MOT, in which the sequence can pause until a desired number of atoms are loaded into the MOT, providing a consistent number of atoms to be loaded into the dipole trap. The resulting fluorescence signal of the MOT exhibits variation at the 0.75% level, corresponding to the same variation in the number of atoms (typically $\pm 150 \times 10^3$). We have also verified the shot-to-shot atom number fluctuation in an optical dipole trap - its level reached 2.6% (corresponds to $\pm 83.2 \times 10^3$ number of atoms), which is still excellent. The increase most likely comes from the somewhat stochastic behavior of the cooling stages following the MOT loading. In the end, with our setup, any process that would introduce more than 2.6% loss with respect to the baseline atom number could be identified even though, in practice, we always aimed at a loss fraction somewhere in the 50%-75% range. Building upon this capability, we then employ the observed trap loss dynamics induced by photoassociation to predict the detectability of additional photoassociation lines. This involves comparing the theoretically calculated photoassociation rates with the experimentally observed trap loss behavior, as delineated by Equation 5.3. Through this method, we can estimate the extent of losses in our trap caused by different transitions and assess whether they would be detectable based on the known variation in atom behavior.

5.3 Spin polarization of ultracold cesium atoms

In the photoassociation experiments described in previous sections, the colliding atoms were in the $F = 4$ hyperfine state (in the MOT) and the $F = 3$ hyperfine state (in the optical dipole trap). However, they were not spin-polarized. The operational principle of a MOT fundamentally limits the ability to achieve spin polarization, but there are no fundamental restrictions in the case of a dipole trap. Consequently, we prioritize the preparation of spin-polarized samples for several reasons. In this thesis, our primary focus is on photoassociation experiments, wherein we aim to enhance the rate of molecule formation through the utilization of Feshbach resonances. The effectiveness of this enhancement is directly influenced by the magnetic field strength cor-

responding to a Feshbach resonance, a factor intricately tied to the spin state of the colliding atoms [26, 31].

The majority of the photoassociation experiments that I have performed relied almost exclusively on cesium atoms confined in an optical dipole trap and spin-polarized in the $F = 3$, $M_F = 3$ state with the setup I have developed for preparing spin-polarized atomic ensembles. However, to enable a much more precise characterization of the experimental setup and to verify the assignment of photoassociation lines, I have also utilized the developed method to prepare cesium spin-polarized in the $F = 3$, $M_F = -3$ state. This method can also be used to prepare samples that have roughly equal populations in spin states $M_F = 3$ and $M_F = 2$, as well as $M_F = -3$ and $M_F = -2$, with a negligible population in the remaining spin states. In the case of polarized bosonic samples, the p -wave collisions are suppressed [207], resulting in the diminishing of rotational lines of $J = 1$ and $J = 3$, and enhancement (quasi-doubling) of the line intensity of $J = 2$ [34]. Thus, samples prepared with mixed spin-state population can be utilized to probe the $J = 1$ and $J = 3$ rotational energy levels, as shown in Figure 5.6.

The preparation of spin-polarized samples is done via a red-detuned beam from the atomic transition of $6s^2S_{1/2}(F = 3) \rightarrow 6p^2P_{3/2}(F' = 2)$. This beam is tailored to have circular polarization (σ^+), with a relatively weak π component generated by the induced tilt on the orientation of the quantization axis, which is essential for manipulating the atomic spin states [32, 33].

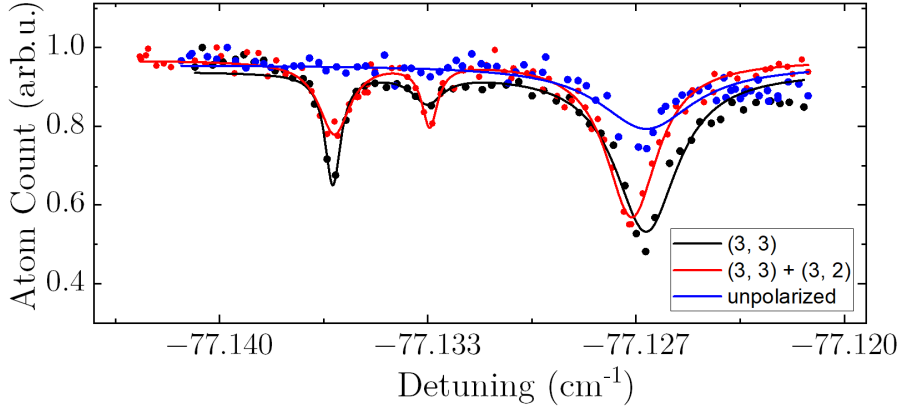


Figure 5.6: The photoassociation spectra of the $\nu = 2$ vibrational level of the $0_g^-(P_{3/2})$ state in the external well, with the unpolarized (blue), polarized to have a roughly equal population in spin states $(F, M_F) = (3, 3)$ and $(3, 2)$ (red), and polarized to $(3, 3)$ (black). The shifts in the spectrum are attributed to the uncertainty of the wavemeter readouts rather than being connected to the induced Zeeman shifts. The detuning is given in reference to the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$.

In the case of driving transitions from $F \rightarrow F' = F - 1$, with circularly polarized light (σ^+), the Zeeman states $M_F = F - 1$ and $M_F = F$ become dark as the σ^+ polarized light cannot excite the atoms populated in these states to F' due to selection rules. As the remaining states are pumped into excited states, these two states in the ground state become populated. However, when π component is added, then the state $M_F = F - 1$ can be pumped to $M'_F = F'$ and consequent decay from this excited state in a cycling transition populates the atoms such that the sample

can be spin-polarized in $M_F = 3$ to a high degree. This process is illustrated in Figure 5.7. For σ^+ transitions, the polarization of the light beam is configured as circular, and the magnetic field is aligned along the k-vector of the beam. To introduce the π component, a slight tilt is applied to the magnetic field. Similarly, atoms can also be populated to $M_F = -3$ by aligning the magnetic field in the opposite direction of the k-vector, with a slight tilt for introducing the π component. Optimal settings for this configuration have been determined and are elaborated upon in the subsequent paragraph.

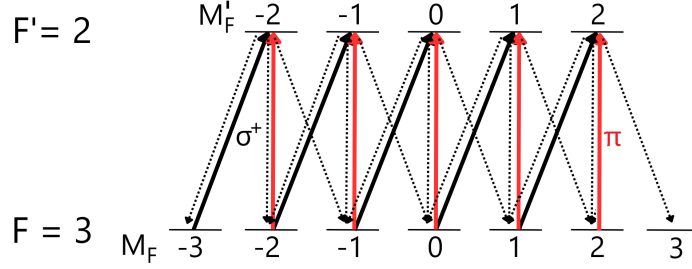


Figure 5.7: Diagram illustrating the spin pumping process of unpolarized cesium atoms in the $F = 3$ state to $F = 3, M_F = 3$ state. The solid black lines represent the pumping to higher Zeeman levels via the σ^+ component of the beam, and the solid red lines represent the π component.

The magnetic field direction was optimized using the following procedure: The starting point for the preparation of spin-polarized samples is a hyperfine-pumped cloud of $F = 3$ cesium atoms confined in the optical dipole trap. The compensation coils are then ramped to produce a magnetic field with the 3 G amplitude to produce a quantization axis. We then turn on the circularly polarized spin-pumping beam for 10 ms.

It should be noted that any operation on the magnetic field used for setting the quantization axis relies, in fact, on controlling currents flowing through three sets of coils in a Helmholtz configuration (see Section 4.3). As such, when we rotate the magnetic field vector, we change, in a correlated way, the currents in compensation coils, which are functions of the applied voltage ($I_{x,y,z} = V_{x,y,z}/R_{x,y,z}$), using the following relations:

$$V_x = \frac{|\vec{B}| \cdot \cos(\theta) \cdot \sin(\phi)}{C_x}, \quad V_y = \frac{|\vec{B}| \cdot \sin(\theta) \cdot \sin(\phi)}{C_y}, \quad V_z = \frac{|\vec{B}| \cdot \cos(\phi)}{C_z}. \quad (5.4)$$

Here, Equation 5.4 represent the voltages to be applied for the x , y , and z coordinates, respectively, corresponding to the Cartesian components of the magnetic field vector $\vec{B}$. $C_{x,y,z}$ are the characterized coefficients of the electromagnetic coils given with units of Gauss per Volt, previously presented in Table 4.2.

To determine the optimal orientation of the applied uniform magnetic field, denoted by angles ϕ and θ , for efficient spin polarization, we initially ramp the magnetic field along these angles. Subsequently, after illuminating the atomic ensemble with spin-polarization light, we confine the atoms in the magnetic trap and monitor the number of atoms in the magnetically trappable $F = 3, M_F < 0$ states (Figure 5.8).

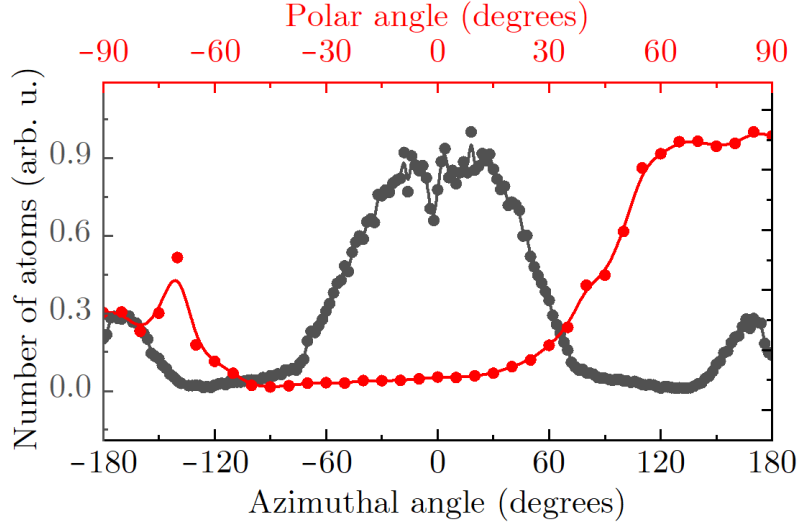


Figure 5.8: The figure illustrates the orientation of the magnetic field during the spin-polarization phase in spherical coordinates, represented by the polar angle (red) and azimuthal angle (black). It shows the atom number trapped in a magnetic trap after spin polarization to the $M_F = -3$ state. The normalized count of atoms represents the number of atoms measured after being held in the magnetic trap for a duration of 300 ms.

Since the remaining atoms in states $F = 3$, $M_F \geq 0$ are not trappable, the optimization of spin polarization to the state $M_F = 3$ can be achieved by identifying the minimum number of trapped atoms. It should be noted that if the direction of the magnetic field vector optimized for polarizing the sample into $M_F = -3$ gets rotated to the opposite direction, we obtain optimal spin polarization in $M_F = 3$. However, thus far, we can only assess the number of atoms pumped to the spin states $M_F > 0$ and relatively to $M_F < 0$ but not individual spin states such as $M_F = 3$ and $M_F = -3$.

To measure the population of atoms in specific spin states, I employed a method that involves the separation of spin states by applying a magnetic field gradient and a uniform bias field. For a given gradient, the force acting on atoms depends on the spin projection M_F on the quantization axis and is described by [138]:

$$F_y = \mu_B M_F g_F \frac{\partial B}{\partial y}. \quad (5.5)$$

For the $F = 3$ state, the Landé factor is $g_F = -1/4$ [45]. Here, μ_B represents the Bohr magneton, and $\frac{\partial B}{\partial y}$ denotes the magnetic field gradient in the vertical direction. In this specific experiment, a uniform magnetic field was applied in the vertical direction in the direction of gravity, along with a magnetic field gradient of 77.805 G/cm, surpassing the required 31.3 G/cm (46.95 G/cm) needed for the magnetic levitation force to compensate the gravitational force for the $F = 3$, $M_F = 3$ ($F = 3$, $M_F = 2$) states. The vertical magnetic field was generated via the rotation of the field used in the spin-pumping process. This rotation was conducted gradually, taking 50 ms to transition, by adjusting the angles θ and ϕ in a linear trend. Rapid reorientation of the magnetic

field direction results in atoms being disturbed and ending up in mixed spin states. The direction of the vertical uniform magnetic field controls the polarity of the magnetic field gradient. The resulting total force acting on atoms in the $M_F = 3$ and $M_F = 2$ states not only opposes gravity but also exceeds its magnitude. Meanwhile, the spin state $M_F = 1$ experiences a lower magnetic levitation force than the gravitational force, causing it to fall downward. However, its downward acceleration is relatively smaller than the acceleration of the $F = 3, M_F = 0$ state, where no magnetic force is exerted. With the magnetic field gradient turned on for an appropriately chosen duration, the spin states become spatially separated. This enables the visualization of spatially distinct spin states, as depicted in Figure 5.9a and Figure 5.9b.

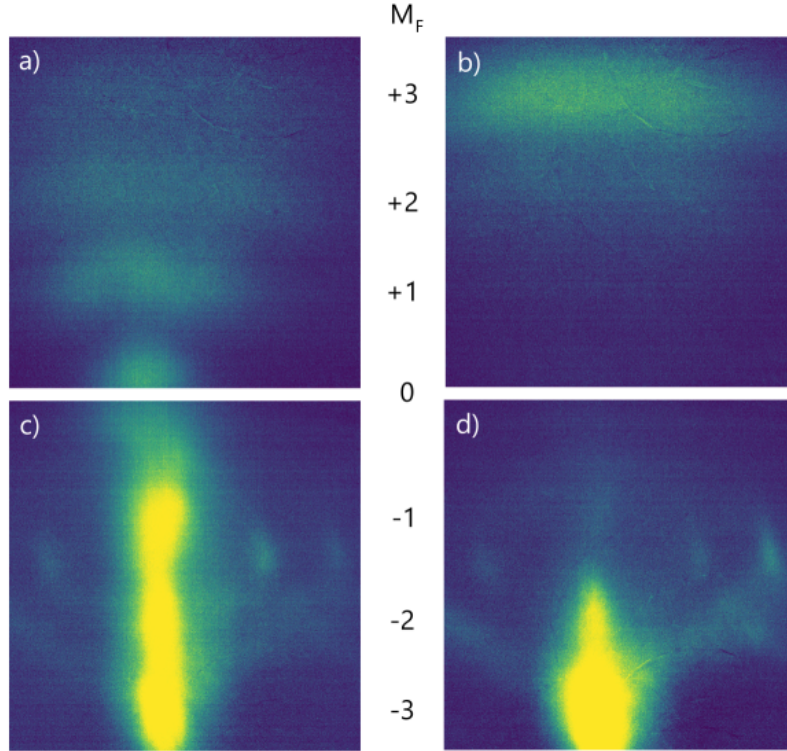


Figure 5.9: Fluorescence readouts of: a) Unpolarized sample showing spin states $F = 3, M_F = 0, 1, 2, 3$. b) Polarized sample to $F = 3, M_F = 3$. c) Unpolarized sample showing spin states $F = 3, M_F = -1, -2, -3$. d) Polarized the sample to $F = 3, M_F = -3$. The images c) and d) are vertically inverted. The center of the magnetic field in all images is towards the bottom.

While theoretically, all spin states could be distinguished within a single experimental sequence, practical constraints emerge due to the limited field of view of the imaging setup. Consequently, achieving spatial separation for $M_F < 0$ states necessitates a slightly modified experimental approach. In this instance, we depart from the previous method of applying a uniform magnetic field opposing gravity, which relocates the magnetic field minima above the sample. This reversal of the magnetic field gradient alters the direction of force exerted on atoms in the $M_F < 0$ states, counteracting the gravitational force. As a result, the spatially distinct spin states can be imaged

within the field of view of the imaging setup, as illustrated in Figure 5.9c and Figure 5.9d.

As one can notice, the spatially distinct spin-states have varying cloud sizes along the horizontal direction. This phenomenon can be attributed to the forces acting on the atoms, encompassing not only the vertical direction but also the horizontal direction due to the presence of a horizontal magnetic field gradient. The horizontal force acting on the atomic cloud can either disperse or condense the atoms depending on the spin state of the individual atoms. We can describe the potential generated by the applied magnetic field for each spin state as [209]:

$$U(x, y, z) = M_F g_F \mu_B |B(x, y, z)|. \quad (5.6)$$

Here, $B(x, y, z) = A\sqrt{4x^2 + (y - \Delta y)^2 + z^2}$ represents a static quadrupole field generated by a pair of electromagnetic coils with opposite currents, where A denotes the gradient along the $y - z$ plane [210]. This setup is complemented by a fixed uniform magnetic field along the y -axis, which shifts the center of the quadrupole field by Δy .

In this context, we present the determined potential along the x -axis (horizontal axis of the images depicted in Figure 5.9), where Figure 5.10 illustrates this potential for all spin-states in the $F = 3$ ground-state level together with the acting force on atoms along the horizontal x -axis. Notably, for higher M_F values, the potential exhibits a steeper gradient, resulting in a greater force thus, faster expansion of the cloud along the horizontal direction compared to other spin states.

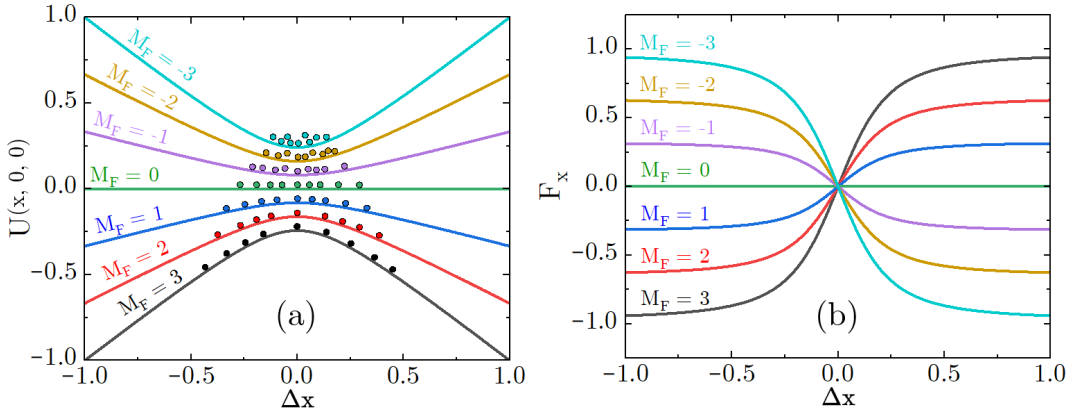


Figure 5.10: (a) Illustration of the trapping potentials $U(x, 0, 0)$ for all possible spin states of $F = 3$ along the displacement from the center of the sample Δx . Only states with $M_F < 0$ are trappable in this configuration, while states with $M_F > 0$ are rejected from the region due to the concave downward potential shape. The points above the potential curves represent a rough estimation of the positions of atoms after a fixed amount of time within the potential. The $M_F = 0$ state only expands due to thermal expansion. (b) Forces acting on the atoms in the horizontal direction for all spin-states of the $F = 3$ ground state versus the position of the atom along the x -axis. All axes in both plots are displayed in arbitrary units.

From these spatially separated readouts of the spin states, the populations corresponding to

different states are determined. The remaining parameters, such as the amplitude of the uniform magnetic field (3.6 G) and the exposure time of the spin-polarization beam (8 ms), are also optimized using this method. These populations are shown in Figure 5.11, where the spin polarization process is studied as a function of the polarizing beam power.

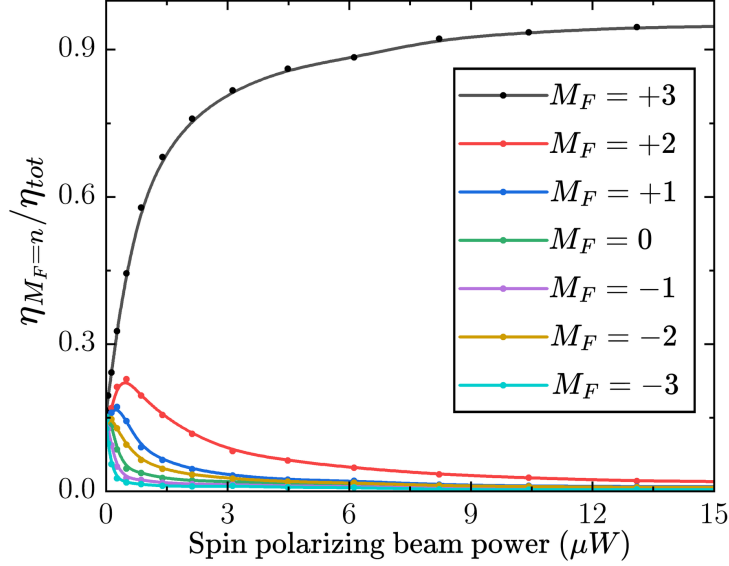


Figure 5.11: The ratio of atoms in each spin state to the total number of atoms is plotted against the power of the spin-polarizing beam. In this experiment, the sample was exposed to light for 8 ms, causing the atoms to be pumped to the $F = 3$, $M_F = 3$ state. The population determination involves a two-step measurement process. Initially, the population of $M_F = 0, 1, 2, 3$ spin states is determined. Subsequently, the remaining spin states are recorded using the same experimental sequence but with a different position of the minimum of the magnetic field, as discussed in this section. Solid lines are provided as guides to the eye.

Our measurements demonstrate that the spin-state polarization sequence can prepare samples in states with $M_F = 3$ or $M_F = -3$ with an efficiency greater than 95%. Simultaneously, we can generate samples with mixed states (achieved only by reducing the power of the spin-pumping beam), with a significant portion of atoms being in the states $M_F = 3$ and $M_F = 2$, or $M_F = -3$ and $M_F = -2$, which is sufficient for studying Feshbach resonances and additional experiments such as photoassociation. Additionally, alternative methods can be applied to deplete atoms in the remaining states by introducing the π polarization component through the tilt on the quantization axis in the later stages of the spin-pumping process, thus selectively populating atoms in states $M_F = 3$ and $M_F = 2$ (or $M_F = -3$ and $M_F = -2$).

After successfully generating spin-polarized atomic cesium ensembles—critical for our objective of utilizing Feshbach-optimized photoassociation—we now aim to maintain their high degree of spin polarization under the generated high magnetic fields. We use a set of coils named ‘Feshbach coils’, designed to produce high-intensity uniform magnetic fields as described in Section 4.3 earlier. For the spin-polarized sample to remain undisturbed by the introduction of another uniform magnetic field, aligning the direction of the quantization axis with the magnetic field generated by the Feshbach coils is necessary.

To verify this alignment, we conduct an experimental sequence as follows. Initially, we load atoms into the optical dipole trap and spin-polarize the sample to the $M_F = 3$ state. Subsequently, we reorient the direction of the quantization axis, ensuring that it is opposite to the estimated direction of the magnetic field generated by the Feshbach coils. Next, we rapidly ramp up the Feshbach coils in a linear fashion up to a certain value and then ramp them down to zero again with a linear trend. Following this, we load the sample into the magnetic trap and monitor the number of trapped atoms.

During this process, we intentionally lower the efficiency of the spin polarization process to ensure the atom cloud remains visible and trackable in each experimental iteration. We anticipate specific outcomes: for the samples prepared to have $M_F = 3$ spin state, we expect an increase in the number of atoms trapped in the magnetic trap due to the rapid change in the quantization axis, leading the atoms to populate spin states $M_F < 0$ (see Figure 5.12). Conversely, for the $M_F = -3$ state, we anticipate a decrease in the number of trapped atoms due to the same manipulation of the quantization axis, increasing the population of atoms in the $M_F > 0$ states.

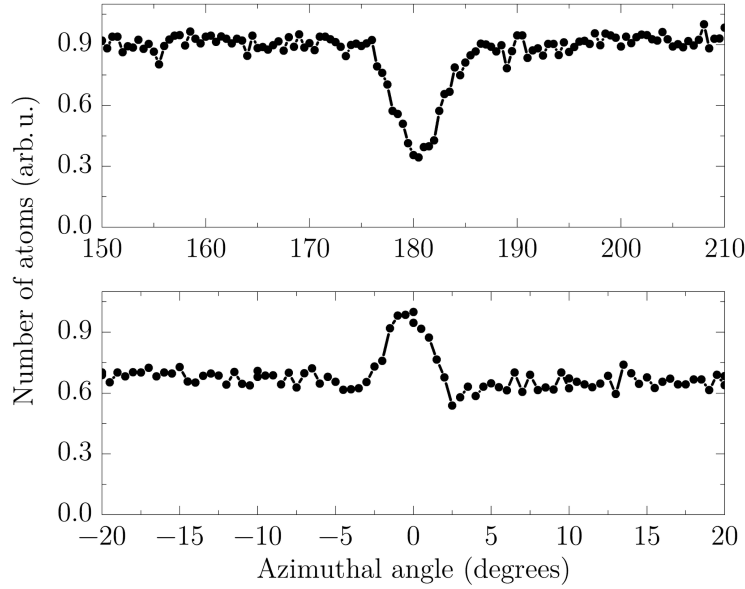


Figure 5.12: The remaining number of atoms in the magnetic trap after ramping the 'Feshbach coils' plotted against the azimuthal (ϕ) angle of the magnetic field applied for spin-polarization of the sample. The lack of atoms in the magnetic trap indicates the spin pumping to $M_F > 0$ states (top), while the increase indicates the spin pumping to $M_F < 0$ (bottom).

5.4 Feshbach optimized photoassociation

In the field of ultracold atomic physics, the manipulation of collisions plays a pivotal role in unlocking control over the interactions between atoms, offering a gateway to diverse applications, including atomic clocks [211], atomic interferometers [212], and Bose-Einstein condensates (BEC) [52].

A detailed description of Feshbach resonances is provided by the comprehensive analysis presented in a review article by Chin et al. [26], and this descriptive section was included under its guidance. A Feshbach resonance is often described via consideration of two molecular potential curves, $V_{bg}(R)$ and $V_c(R)$. Here, V_c has lower potential energy than $V_{bg}(R)$ in any given internuclear distance R . As the internuclear distance (R) becomes large, the molecular potential $V_{bg}(R)$ approaches the potential energy of two free atoms in an ultracold sample. In a collision process of two atoms, the $V_{bg}(R)$ potential is referred to as the entrance channel, while the V_c is a closed channel in which V_c can have bound molecular states near the threshold of the open channel. A Feshbach resonance is observed when the bound molecular state in the closed channel comes close in energy to the scattering state in the open channel. Even with weak coupling, significant mixing between these two channels can occur. The variation in energy can be manipulated by adjusting a magnetic field when the associated magnetic moments are distinct, resulting in a Feshbach resonance that is magnetically tuned [213, 214]. In this case, the scattering length, $a(B)$, can be described in terms of the present magnetic field [215]:

$$a(B) = a_{bg} \left(1 - \frac{\Delta}{B - B_0} \right), \quad (5.7)$$

where, a_{bg} is the scattering length related to the V_{bg} molecular potential, and it is also related to the last bound vibrational level of this potential. The Δ is the width of the resonance, and B_0 is the magnetic field position of the resonance. When the condition of $B = B_0 + \Delta$ is satisfied, the scattering length reaches zero, commonly referred to as the ‘zero crossing’.

Practically, Feshbach resonances have played a crucial role in achieving significant milestones, including the realization of Bose-Einstein condensation in ultracold ^{133}Cs [52, 216] and ^{39}K atoms [217]. These accomplishments marked the beginning of a series of similar experiments with cold atoms, revealing the versatility of Feshbach resonances in various atomic systems. In particular, Feshbach resonances (FR) have played a pivotal role in condensing atoms with negative scattering lengths at zero magnetic field, providing the capability to tune the scattering length to positive values—an essential feature for Cs [218, 219], ^{39}K [220, 99], ^7Li [221], and ^{85}Rb [222]. Notable applications include the formation of solitons [223], accessing the BEC-BCS crossover in fermions [224, 225, 226], the observation of Efimov states [227], the formation of loosely bound Feshbach molecules in both homonuclear and heteronuclear configurations [228, 229, 230], the collapse of a Bose-Einstein condensate known as Bose-nova [231], and the evaporation of fermions [232]. While numerous examples exist, these represent some of the most prominent applications in the field.

The concept of ‘Feshbach-optimized photoassociation’ (FOPA) is a significant advancement, offering a new approach to use Feshbach resonances for improved control of photoassociation processes [27, 28, 29, 30]. FOPA utilizes the unique features of Feshbach resonances to modify scattering cross-sections, influencing the association and dissociation of atoms during photoassociation. This technique shows potential in refining the precision and efficiency of photoassociation spectroscopy. The photoassociation process relies on atoms colliding with each other; thus, the ability to tune the collisional properties yields the enhancement of photoassociation rates under certain circumstances [34]. However, the initial properties of the atoms must be well known, and their energy states should be well controlled.

With successfully spin-polarized Cs samples, the investigation transitioned to exploring Feshbach resonances in polarized Cs atoms. This exploration was undertaken by consideration of the

potential to use Feshbach resonances to enhance the photoassociation rate of Cs_2 molecules.

In our single-chamber setup, four sets of coils were installed to generate a uniform magnetic field (refer to Section 4.3). These coils were positioned so that each provided a uniform magnetic field along a specific axis. The initial three sets compensated for external magnetic fields and facilitated the movement of the ultracold sample in the presence of a magnetic gradient. These coils are intended for generating lower magnetic fields, while the fourth set was dedicated to generating high, uniform magnetic fields along the x -axis, specifically designed for Feshbach measurements. This fourth set can safely generate a magnetic field of up to 300 G without the need for additional cooling methods. Calibration of the fourth set (Feshbach coils) was performed by referencing it to the previously calibrated three sets of coils.

Subsequently, Feshbach measurements in the optical lattice were performed, and the results demonstrated a significant correlation with previously published works [162, 31]. Each Feshbach resonance detected through our setup had been observed in these prior studies. In these measurements, the atoms were loaded into the optical lattice facilitated via the dipole trapping laser, and then the atoms were spin-polarized into $F = 3$, $M_F = 3$ state. However, few of the Feshbach resonances were observed, and certain Feshbach resonances initially were not detected. To address this challenge, a method exploiting the enhancement of the photoassociation rate through Feshbach resonances was employed, as detailed in [233]. Initially, the laser frequency is set to a specific frequency, inducing a loss of atoms through photoassociation. Subsequently, adjusting the magnetic field leads to an increase in photoassociation-induced loss, particularly as the magnetic field approaches a Feshbach resonance. In this context, the sample was illuminated with a photoassociation laser tuned to the frequency of a transition to a known photoassociation level namely $\nu' = 235$ of $1_g(\text{P}_{3/2})$, and by setting the magnetic field to a Feshbach for a duration of ~ 1.5 seconds, Feshbach resonance enhanced photoassociation was simply triggered (see Figure 5.13).

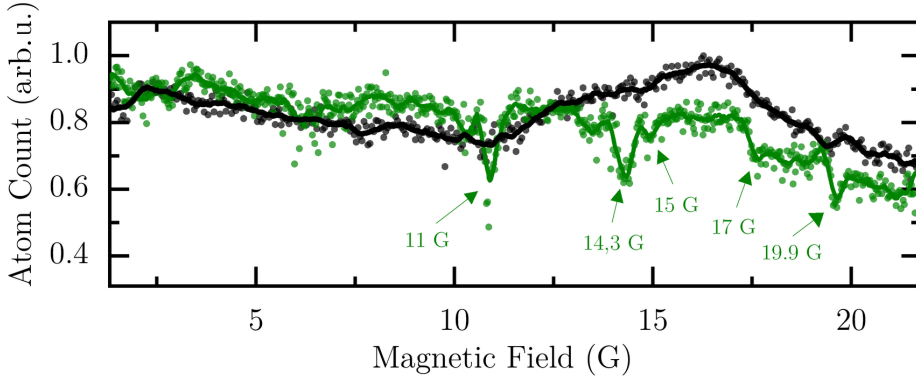


Figure 5.13: Observed Feshbach resonances up to 22 G for atoms in the state $(F, M_F) + (F, M_F) = (3, 3) + (3, 3)$ (shown by the black line) are obtained, relying on collisions and three-body losses for observing Feshbach resonances. The green line represents the results obtained when the atomic sample is illuminated with light resonant with a transition from a pair of two free atoms to an excited molecular level, namely $\nu' = 235$ of $1_g(\text{P}_{3/2})$.

This enhancement is explained in detail in the Ref. [34]. Briefly, the photoassociation rate Γ_{PA}

is approximately proportional to the square of the integral overlap, commonly known as the Franck-Condon factor, between the radial wave function Ψ_ν of the excited state and the wave function Ψ_E of two atoms with collision energy E . The expression of the photoassociation rate can be simplified to

$$\Gamma_{\text{PA}} \propto |\Psi_E(R_+(\nu))|^2, \quad (5.8)$$

where, $R_+(\nu)$ represents the outer turning point of the excited rovibrational level ν , J . Essentially, photoassociation towards a specific excited rovibrational molecular level occurs at a particular internuclear distance. Consequently, the photoassociation efficiency is closely linked to the probability of the presence of two atoms at a given distance $R_+(\nu)$. By varying the magnetic field value, one can significantly change the collisional properties, as the scattering length varies with the applied magnetic field. This variation increases the likelihood of two atoms being present at a desired distance from each other. The scattering length value under applied external magnetic fields mostly varies smoothly and corresponds to a very broad range of magnetic fields. In this context, Feshbach resonances, characterized by a steep and substantial variation of the scattering length (as shown in Equation 5.7), can be utilized as a tool for enhancing the photoassociation rate.

This approach facilitated the detection of Feshbach resonances previously undetected in our setup, and the corresponding magnetic field values were utilized in subsequent experiments to enhance the photoassociation of the two lowest vibrational levels in the external well of the $0_g^-(P_{3/2})$ state of Cs_2 and to extend the spectroscopic data related to the states $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$.

5.5 Photoassociation spectroscopy of 0_u^+ and 1_g states of Cs_2 converging to the $6S_{1/2} + 6P_{3/2}$ asymptote.

After the improvements in our photoassociation setup, namely for photoassociation of Cs_2 , improvements in the imaging system, improvements in the general stability of the loaded atoms into the dipole trap via the implementation of the conditional loading method, and most importantly enhancing photoassociation by Feshbach resonances with spin-polarized samples have given us notable advancements in measuring photoassociation lines. In this section, the experimentally observed photoassociation transitions to the $0_u^+(P_{3/2})$ state and the $1_g(P_{3/2})$ state are presented.

The experiments for these states are conducted in an optical dipole lattice, where the local densities of the ultracold sample are higher than in an optical dipole trap, thus increasing the photoassociation rate. Photoassociation lines were detected using a trap-loss spectroscopy method with a 1.5-second exposure time for photoassociation. The photoassociation beam power was maintained at a constant 15 mW, and an external magnetic field of 47.9 G, targeting a specific Feshbach resonance, was applied during this exposure. The loaded atoms were spin-polarized to the $F = 3$, $M_F = 3$ state. Through this sequence, numerous vibrational levels belonging to the $0_u^+(P_{3/2})$ state and the $1_g(P_{3/2})$ state were probed. The corresponding energies of the probed vibrational levels for the $0_u^+(P_{3/2})$ state are presented in Table 5.1, while those for the $1_g(P_{3/2})$ state are shown in Figure 5.14.

Table 5.1: The experimentally observed photoassociation transitions to levels of the $0_u^+(P_{3/2})$ state converging to $6S_{1/2} + 6P_{3/2}$ limit. The level marked with an asterisk is the level previously reported in the [25]. Their vibrational levels are labeled with $\nu' = [\nu_D - \nu]$, where ν_D denotes the vibrational level lying closest to the dissociation limit. The detunings are referred to the $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$ atomic transition. The vibrational levels beyond $\nu' = 257$ have been assigned numerical labels based on the results of numerical fits; their absolute vibrational numbers may differ from the actual case. The numerical fitting process and the numbering methodology are discussed further in the section.

ν'	Δ (cm^{-1})	ν'	Δ (cm^{-1})
233*	-68.14703	249*	-102.23688
234*	-69.95135	250*	-104.65975
235*	-71.75137	251*	-107.16774
236*	-73.5808	252*	-109.71044
237*	-75.67519	253	-112.29185
238*	-77.54302	254	-114.75243
239*	-79.5009	255	-118.29959
240*	-81.50074	256	-120.46347
241*	-83.53292	257	-122.64256
242*	-85.63642	269	-163.08201
243*	-87.65712	276	-190.08464
244*	-89.64246	284	-225.85926
245*	-91.43861	286	-233.71442
246*	-95.03233	291	-260.83948
247*	-97.6534	296	-289.48495
248*	-99.85863		

Table 5.2: The experimentally observed photoassociation lines for transitions to levels of the 1_g state converging to $6S_{1/2} + 6P_{3/2}$ limit. The level marked with an asterisk is previously reported in the Ref. [25]. Their vibrational levels are labeled with $\nu' = [\nu_D - \nu]$, where ν_D denotes the vibrational level lying closest to the dissociation limit. The detunings are referred to the $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$ atomic transition. The vibrational levels beyond $\nu' = 252$ have been assigned numerical labels based on the results of numerical fits; their absolute vibrational numbers may differ from the actual case. The numerical fitting process and the numbering methodology are discussed further in the section.

ν'	Δ (cm^{-1})	ν'	Δ (cm^{-1})
229*	-68.5151	246	-105.86829
230	-70.37215	247	-108.46174
231	-72.21156	248	-111.21822
232	-74.13866	249	-113.966
233	-76.04054	250	-116.69733
234	-78.08561	251	-119.67153
235	-80.1328	252	-122.64256
236	-82.2099	263	-159.63294
237	-84.33976	270	-187.98599
238	-86.51363	278	-225.27213
239	-88.76959	279	-229.87135
240	-91.02632	280	-234.80862
241	-93.30175	284	-258.14353
242	-95.77236	289	-285.53353
243	-98.19537	290	-293.97536
244	-100.68612		
245	-103.19333		

It should be noted that, in fact, the main motivation of this work has never been to measure the photoassociation spectra of the $0_u^+(P_{3/2})$ and the $1_g(P_{3/2})$ states but to benchmark the photoassociation capabilities of the developed setup. Therefore, the intention was not to measure all vibrational levels of these states but to determine the capabilities of our setup by trying to find the photoassociation level with the lowest intensity that can be detected. In the previously published scientific papers focusing on these levels, both states were extended to detunings up to -112 cm^{-1} from the atomic asymptote [25], which are indicated with asterisks in Table 5.1 and in Table 5.2. In this study, we extended the range up to detunings of -300 cm^{-1} , marking a great benchmark for our experimental setup.

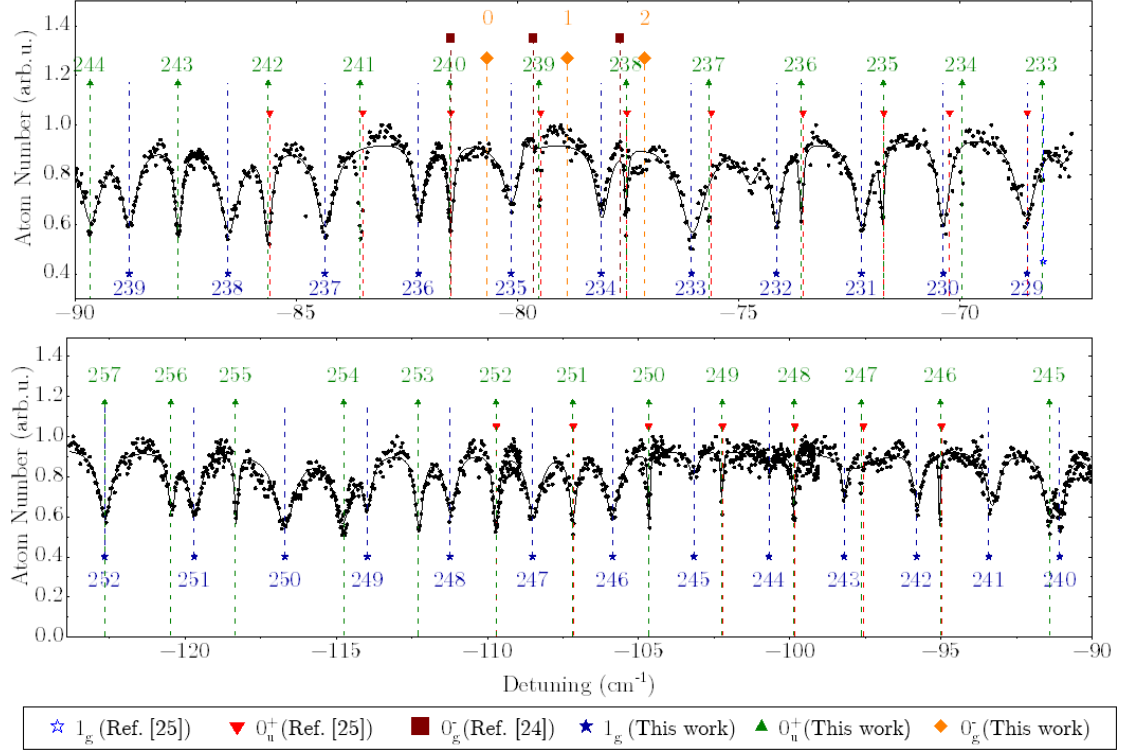


Figure 5.14: Photoassociation spectra of the states $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. The detunings are referred to frequency of the $6s^2S_{1/2}(F=4) \rightarrow 6p^2P_{3/2}(F'=5)$ atomic transition. The measured three lowest vibrational levels in the external well of the state $0_g^-(P_{3/2})$ are shown. The vertical dashed lines represent data reported from the Refs. [24, 25] and present work.

The spectrum presented in Figure 5.15 corresponds to transitions involving the last two vibrational levels (which can be observed with our experimental setup) of the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. These vibrational levels represent the outermost limits measurable by our experimental setup. Specifically, these levels were probed using the $B = 47.9 \text{ G}$ Feshbach resonance with $F = 3$ and $M_F = 3$ to enhance the photoassociation rate. Despite the rigorous efforts to measure further photoassociation lines done with Feshbach resonances other than 47.9 G (below 60 G), these attempts remained unsuccessful.

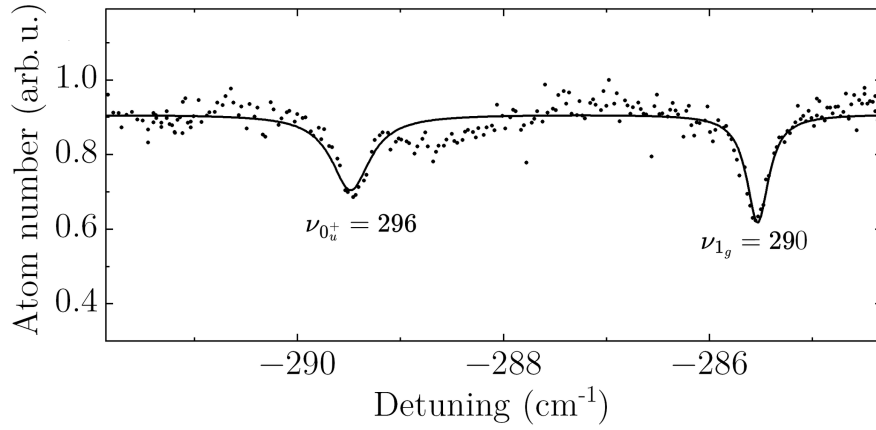


Figure 5.15: Fragment of the photoassociation spectra of the $0_u^+(P_{3/2})$ ($\nu' = 295$) and the $1_g(P_{3/2})$ ($\nu' = 290$). These are the lowest vibrational levels of both states observed in our experiments, which have the largest detuning from the dissociation limit. The detuning is given in reference to the atomic transition $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$.

The vibrational levels we have probed exhibited rotational unresolved for both states, with their observed widths typically being approximately 8-10 GHz, indicating significant broadening. This broadening likely stems from the predissociation of the $1_g(P_{3/2})$ and $0_u^+(P_{3/2})$ states, as proposed by Pichler et al. (2006) [109].

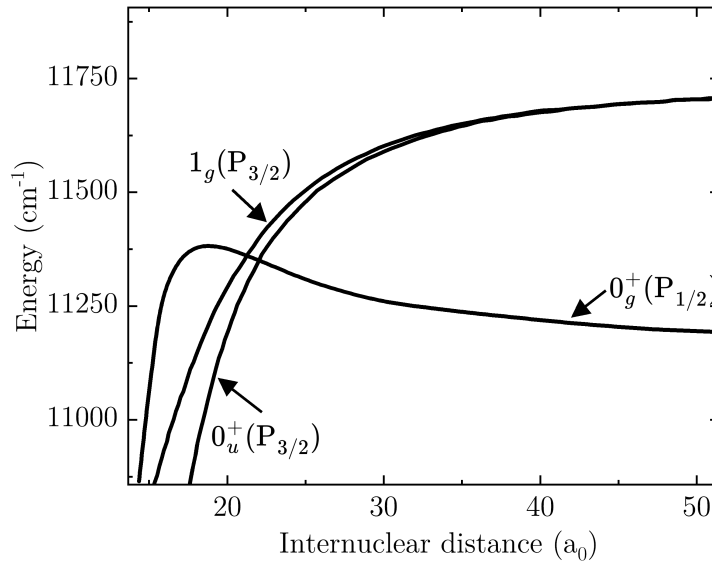


Figure 5.16: Scheme of selected molecular potentials of Cs_2 suggesting possible predissociation of the $1_g(P_{3/2})$ states via crossing with the $0_g^+(P_{1/2})$ potential. The $0_u^+(P_{3/2})$ state predissociates above the $6S_{1/2} + 6P_{1/2}$ atomic asymptote. The potential energy curves are taken via digitization from Refs. [22, 109].

The predissociation mechanism of the $1_g(P_{3/2})$ state can be explained through a crossing event with the repulsive $0_g^+(P_{1/2})$ state at approximately $22a_0$ [109]. Below $20a_0$, the $0_g^+(P_{1/2})$ state transitions to an attractive state due to an avoided crossing with the $0_g^+(P_{3/2})$ state. This transition results in a second crossing with the $1_g(P_{3/2})$ state at around $16.8a_0$ as depicted in Figure 5.16, contributing to a more complex predissociation process. Notably, our measurements extended to detunings of -300 cm^{-1} , allowing us to probe vibrational levels at internuclear distances much closer to the crossing point between the states $1_g(P_{3/2})$ and $0_g^+(P_{1/2})$ which corresponds to a detuning of -366.18 cm^{-1} .

On the other hand, when the predissociation of the $0_u^+(P_{3/2})$ state is concerned, two electronic states $A^1\Sigma_u^+$ and $b^3\Pi_u$, contribute to two 0_u^+ states converging to the $6S_{1/2} + 6P_{1/2}$ and $6S_{1/2} + 6P_{3/2}$ asymptotes. The strong spin-orbit interaction, especially pronounced in cesium, leads to the mutual interaction of these states, which is responsible for the observed predissociation of $0_u^+(P_{3/2})$. This process resembles a ‘half collision’ with a fine-structure change, similar to extensively studied collisions [109, 234, 235, 236, 237] involving fine-structure change. Previous efforts by Bai et al. [238] and Znotis et al. [239] aimed to deperturb these states using experimental data on low and medium excited rovibrational levels. However, their analysis presented two drawbacks: the resulting potential energy curves had an incorrect analytical form in the asymptotic region and yielded inaccurate asymptotic positions.

Following our novel observations, we attempted to fit the obtained experimental data to existing molecular models describing the potentials of these states in long-range region. The long-range parts of potential energy curves of the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states of the Cs_2 molecule are often described with the improved LeRoy-Bernstein near-dissociation expansions [240, 241]. The calculation of long-range limits of alkali-metal dimer molecular states which dissociate to the atom pairs $S_{1/2}$ and $P_{3/2}$ is given by using almost degenerate perturbation theory [112]. In the long-range region, the experimental data obtained for these states close to the dissociation limit can be expressed in terms of energies of the rovibrational levels [25, 242, 240, 241]:

$$E(\nu, J) = E_D + \sum_{j=0}^{\infty} Q_j \left(\frac{\gamma_D - \gamma}{\gamma_D} \right) X_j(n) \times C_n^{-2/(n-2)} (\gamma_D - \gamma)^{4/(n-2)-2(j-1)} \lambda^j. \quad (5.9)$$

In the above equation, the E_D is the dissociation energy, and C_n is the leading dispersion coefficient as given in Table 3.1. The remaining variables are determined by the vibrational number (ν) and rotational number (J) of the corresponding energy level, and λ , γ , and γ_D are defined as the following:

$$\lambda = G^2(m)[J(J+1) - \delta], \quad (5.10)$$

$$\gamma = G(m)(\nu + 1/2), \quad (5.11)$$

$$\gamma_D = G(m)(\nu_D + 1/2). \quad (5.12)$$

Here, $G(m)$ is $\hbar/\sqrt{2m}$, and m is the reduced mass of the system. The δ is defined as Ω^2 for the

Hund's case (c). The values of $X_j(n)$ and Q_j are given in Ref. [25] for various values of j and n . ν_D is the effective vibrational quantum number at the dissociation limit.

For the analysis of the data obtained in the experiments for the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states, Equation 5.9 is employed to determine the dispersion coefficients by having a standard least-squares fitting method [25]. Nevertheless, upon encountering detunings below -120 cm^{-1} , both states exhibited unexpectedly high residuals in attempts to find the optimized best fit of Equation 5.9. When computing the vibrational energy levels with parameters derived from the optimal potential proposed by Ma et al. [25], we once again encountered consistently high error values for vibrational levels with higher detunings. The calculated residuals corresponding to the vibrational levels of the $1_g(P_{3/2})$ state are presented in Figure 5.17.

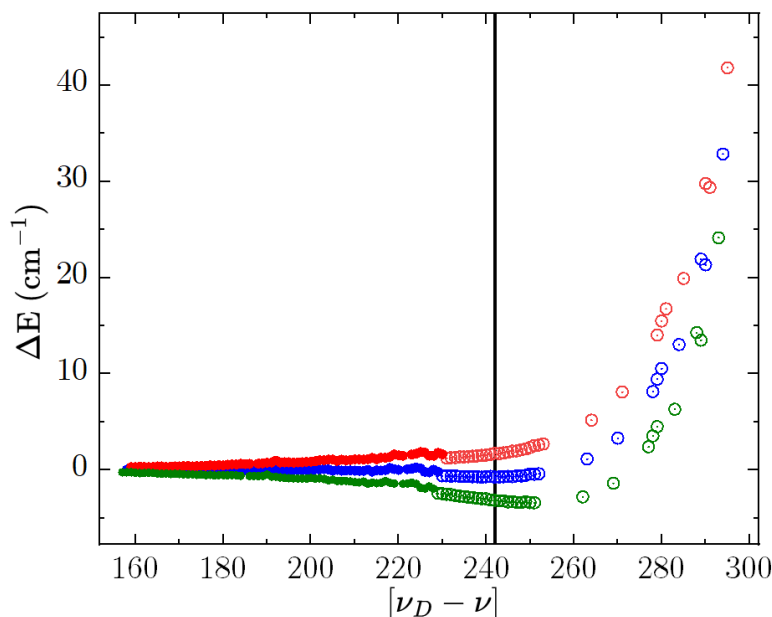


Figure 5.17: Comparison of vibrational energy residuals determined for the vibrational energy levels of $1_g(P_{3/2})$ state by the best fitting parameters obtained in the Ref. [25]. The figure plotted with respect to vibrational numbering $\nu' = [\nu_D - \nu]$. Blue data points represent the best fitting where the corresponding residual is minimum. Green data points represent the case where $\nu = \nu_{best} + 1$ and red data points represent $\nu = \nu_{best} - 1$. Solid data points are obtained by the experimental results disclosed in Ref. [25]. The vertical line corresponds to the vibrational number at which the outer turning point of the vibrational wavefunction falls below the modified LeRoy radius.

This inconsistency raised a critical discussion regarding the appropriateness of the employed theoretical approach. Specifically, the combination of LeRoy–Bernstein near-dissociation theory [240] and the almost degenerate perturbation theory by Marinescu and Dalgarno [243, 112] may be encountering limitations below certain outer turning point R_{v+} , suggesting the need for a more refined theoretical framework.

In contrast, Liu et al. [244] adopted Hund's case (c) basis, suitable for long-range analysis,

where an avoided crossing of $0_u^+(P_{1/2})$ and $0_u^+(P_{3/2})$ states was considered, explicitly accounting for spin-orbit interaction. Leveraging experimental data provided by Bai [238] and additional insights from Ma et al. [25], Liu et al. constructed numerical potentials for both 0_u^+ states, ensuring accurate asymptotic behavior. By employing the potential energy curve specifically for the $0_u^+(P_{3/2})$ state, we determine the absolute vibrational quantum numbers for the observed vibrational levels in our present experiment through the solution of the radial Schrödinger equation. The residuals are then obtained by comparing the calculated energies with experimentally measured values derived from the present work. While the vibrational energy levels observed by Liu et al. [244] and this work exhibit satisfactory agreement for the higher levels, a discrepancy of approximately 0.2 cm^{-1} arises due to an inaccurately positioned asymptote [245]. Although the potential of Liu et al. seemingly allows the assignment of absolute vibrational quantum numbers to photoassociation-observed levels from their experiment, it reveals discrepancies for lower vibrational levels obtained from our experiment, as shown in Figure 5.18, however, the numbering of these levels seems to be unambiguous.

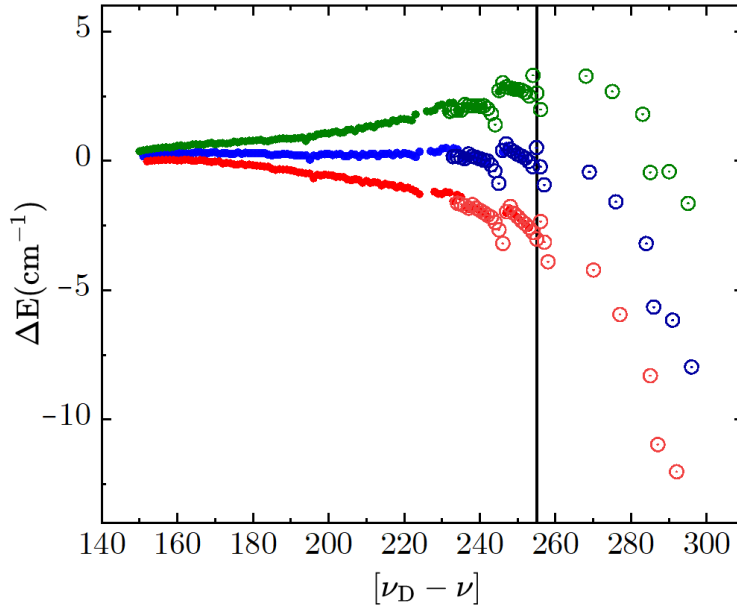


Figure 5.18: Comparison of vibrational energy residuals determined for the vibrational energy levels of $0_u^+(P_{3/2})$ state. The figure plotted with respect to vibrational numbering $\nu' = [\nu_D - \nu]$ corresponding to vibrational numbering ν_{best} of the levels observed in the present experiment and in Ref. [25]. Blue data points represent the best fitting where the corresponding residual is minimum. Green data points represent the case where $\nu = \nu_{best} + 1$ and red data points represent $\nu = \nu_{best} - 1$. The vertical line corresponds to the vibrational number at which the outer turning point of the vibrational wavefunction falls below the modified LeRoy radius. Solid data points are obtained by the experimental results disclosed in Ref. [25].

The attempt to refit the potential energy curve by fixing the bottom to the potential energy curve provided in Ref. [244] and fitting the upper part to all levels observed in photoassociation experiments proved to be impossible without having at least some data covering the intermediate range of energies. Any modification to the potential would change the numbering of the

uppermost levels in an unpredictable way. The highest experimental level included in the analysis by Znotins et al. [239], corresponds to $\nu' = 394$, while the lowest level observed in our experiments would be $\nu' = 296$. Therefore, approximately 98 vibrational levels are missing. (see Figure 5.19), preventing accurate modification of the potential without altering the numbering of upper levels unpredictably. Moreover, the failure of Liu et al.'s potential to reproduce correct energies of lower vibrational levels observed by them can be attributed to the limitations of their asymptotic expansion, valid only beyond the modified LeRoy radius (17.6 Å) [111].

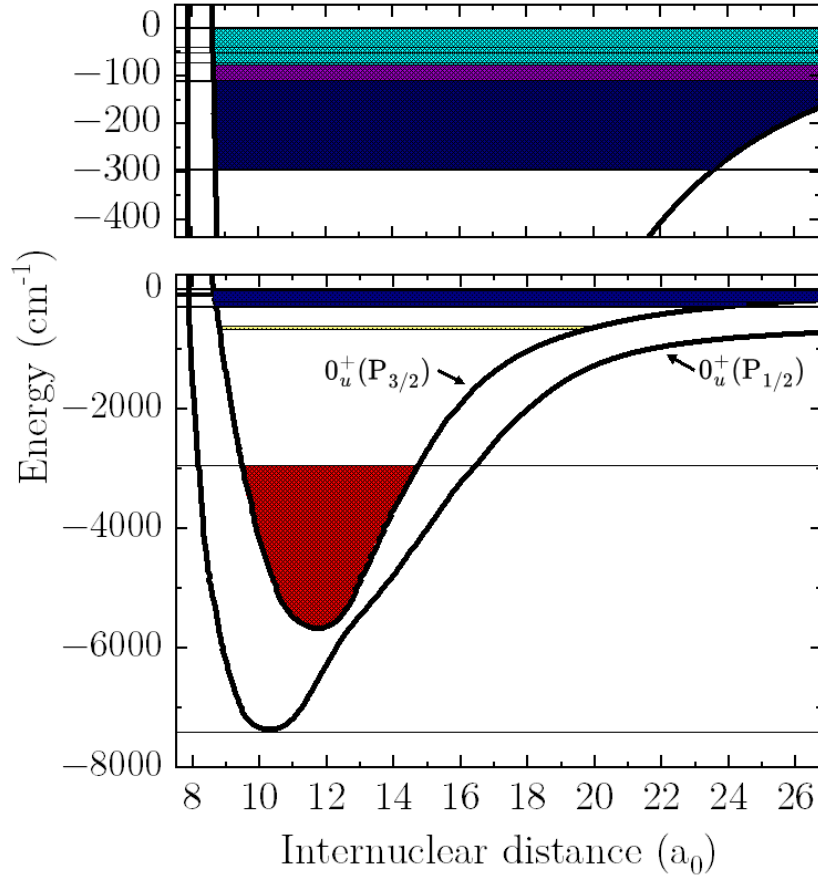


Figure 5.19: The potential energy curves of the $0_u^+(P_{1/2})$ and $0_u^+(P_{3/2})$ states. The energy is referenced to the $6S_{1/2} + 6P_{3/2}$ asymptote. The color patterns represent data from previous experiments: Ref. [37], Ref. [42], and Ref. [43] (cyan) cover the range above the $6S_{1/2} + 6P_{1/2}$ asymptote, while Ref. [25] (purple) and our work (dark blue) also cover this range. Ref. [246] (yellow) and Ref. [246, 238] (red) cover the range below the $6S_{1/2} + 6P_{1/2}$ asymptote.

In summary, the accepted 'best' absolute numbering aligns with Liu et al.'s potential energy curve, demonstrating its validity for levels within 120 cm^{-1} from the atomic asymptote (corresponding to $\nu' \leq 255$ for $0_u^+(P_{3/2})$ state and due to the same reasoning $\nu' \leq 242$ for the $1_g(P_{3/2})$ state). However, it falls short for levels more distant from the $6S_{1/2} + 6P_{3/2}$ asymptote.

5.6 High resolution photoassociation spectroscopy of 0_g^- state of Cs_2 converging to the $6S_{1/2} + 6P_{3/2}$ asymptote.

The primary focus of this thesis is the external potential well of the 0_g^- state converging to the $6S_{1/2} + 6P_{3/2}$ limit (shown in Figure 5.20). In particular, the external well of the $0_g^- (P_{3/2})$ state has emerged as a significant benchmarking tool for our constructed photoassociation setup.

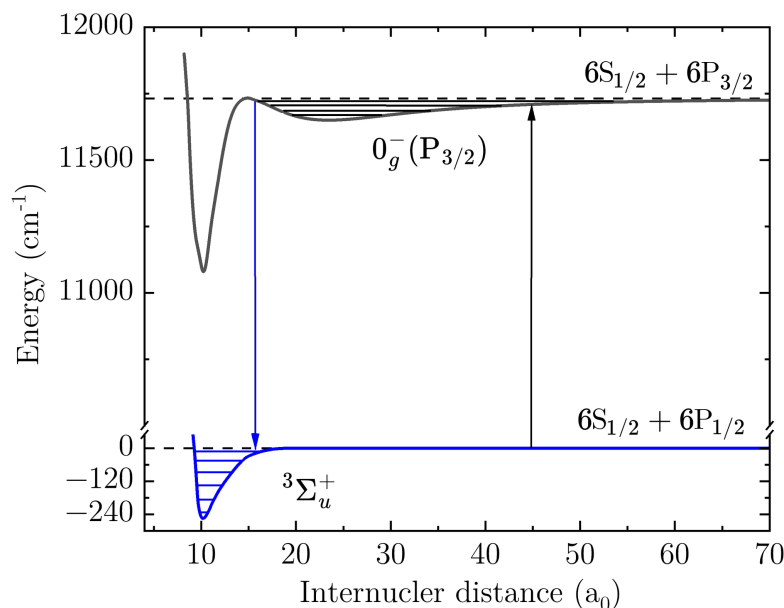


Figure 5.20: Schematic view of the excitation and decay processes of the 0_g^- state of Cs_2 dissociating towards the $6S_{1/2} + 6P_{3/2}$ limit.

This state has been thoroughly investigated by several research groups. Analyses of the potential energy curve, derived from experimental data [21], have revealed certain inconsistencies. These discrepancies were subsequently addressed and resolved by Bouloufa et al. [23]. In the latter study, the experimental data obtained from photoassociation spectroscopy was analyzed via the intensity variation of the spectrum of the Cs_2^+ ion yield, serving as a direct manifestation of the nodal structure of the colliding atom wave functions. Thus, by analyzing the photoassociation rate, they were able to extract the scattering length and the C_6 dispersion coefficient of the lowest triplet molecular state. However, when these resulting values were compared to the results of other experiments done via two-color photoassociation [247] and a trustworthy value obtained at NIST by using magnetically induced Feshbach resonances [162], the values seemed to disagree with the analysis done via the photoassociation spectroscopy. Thus, the theoretical fitting model was re-examined within the same reported work by Bouloufa et al. [23].

The revised theoretical model presented after the reexamination has once again produced results inconsistent with the discrepancies observed in previous measurements. This has led to a suggestion that the vibrational numbering was done incorrectly and that the depth of the external well was possibly deeper than initially anticipated. Essentially, resolving the current disparities could entail increasing the vibrational numbering by two, thereby reaching a conclusion that suggests

the presence of two previously unobserved vibrational levels lying at the bottom of the external well. Despite extensive study over the past few years, these ‘missing’ vibrational energy levels remained unobserved.

The inability to probe these vibrational levels in previously reported experiments could be attributed to their exceptionally low molecular formation rates. These ‘missing’ vibrational levels were expected to display low Franck–Condon factors due to a very small overlap between the excited state and the ground state wave functions. The intensity of the relevant transitions to vibrational levels was reported as a fraction of the intensity of the transition to the vibrational level previously believed to be the vibrational ground state, $\nu = 2$, as 0.8 for $\nu = 1$ and 10^{-3} for $\nu = 0$.

The ‘missing’ vibrational levels were initially reported to be observed by Zhang et al. [24] within a setup comprising an MOT. This observation was made using cesium samples prepared with densities significantly lower than those achieved in the context of photoassociation spectroscopy in optical dipole traps. At the same time, the energies reported in this particular paper were incorrect for the previously known $\nu = 2$ vibrational energy level, with a substantial deviation of 0.42 cm^{-1} (12.6 GHz). This discrepancy, notably higher than the hyperfine splitting of cesium atoms in the ground state, was attributed to a calibration error arising from differences in the calibration benchmarks of the photoassociation laser frequency. The presence of such a significant calibration error in experimental work raises doubts about the validity of the reported energies for the remaining levels, particularly those pertaining to $\nu = 0$ and $\nu = 1$.

The discrepancies observed in both theoretical and experimental studies of the $0_g^-(P_{3/2})$ state have prompted our attention and motivated us to conduct benchmarking experiments focusing on this specific state. Simultaneously, the expected extremely low molecular formation rates align with our efforts to enhance our photoassociation setup by implementing previously known methods, which are crucial for our future experiments planned on potassium-cesium mixtures. Our photoassociation setup, together with the experiments detailed in the previous sections has led to the probing of levels reported in Ref. [24] and assumed to be the ‘missing’ ones; however, it should be noted that when investigated carefully, it became obvious that these levels actually belonged to the $0_u^+(P_{3/2})$ state and not to $0_g^-(P_{3/2})$ state. Since then, I have dedicated a tremendous amount of time to improving our photoassociation setup and, in general, our cesium cooling methods to be able to observe the actual two ‘missing’ vibrational levels. Finally, upon successive enhancements of the photoassociation rate, the two lowest-lying vibrational levels were measured.

Our photoassociation measurements, which probed the lowest-lying vibrational levels, were conducted in a 1D optical lattice. The cesium atoms are loaded into an optical lattice within a single chamber MOT, where the MOT loading times are controlled by tracking the fluorescence signal of the trapped cesium atoms. By doing so, it was ensured that the prepared sample would have a consistent number of atoms in each iteration of the sequence. Once the cesium atoms were loaded into the optical lattice, they were spin-polarized to the state $F = 3$, $M_F = 3$ by utilizing a σ^+ polarized laser beam with a small portion of π component as well. The spin-polarization beam was blue-detuned by 8 MHz from the $F = 3$ to $F' = 2$ transition. The spin-state polarization stage has the efficiency of $\eta_{spin} \geq 95\%$. The number of atoms loaded into the optical lattice is typically around 3.2×10^6 atoms with the temperature of $\sim 12 \text{ } \mu\text{K}$. In the context of photoassociation measurements, the optical lattice yields higher densities when compared to our optical dipole trap. Before the photoassociation beam was illuminated onto the prepared sample,

an external uniform magnetic field was ramped to 19.7 G (Feshbach resonance line of cesium for $F = 3$, $M_F = 3$ state) [162]. The tunability of the radial variation of the collisional wave function with an external magnetic field results in a significant enhancement of the photoassociation rate [34].

As the photoassociation laser, I have utilized the commercially available Ti:Sapphire laser (MSquared - SolsTiS-SRX), the installation and light preparation setup of which were introduced in Section 4.6. A small portion of the photoassociation beam is used for optical frequency referencing to the frequency comb. A fraction of this light is coupled directly into a wavemeter (WS-7) for a rough readout of its frequency with the absolute precision of ≤ 60 MHz. The remaining fraction is then overlapped with the beam of a custom-made tunable frequency comb (830 nm to 880 nm) for beat note based optical locking. Once the frequency of the photoassociation laser is locked to one of the teeth of the frequency comb, by changing the repetition rate of the frequency comb, the photoassociation laser frequency is tuned to the desired value (as formulated with Equation 4.32). The resulting absolute precision of the photoassociation laser frequency was ≤ 8.87 kHz with its line-width ≤ 50 kHz. The photoassociation beam is aligned to pass through the entire sample along its elongated axis, as the cloud exhibits a cylindrical symmetric Gaussian-like density distribution. The beam is focused at the center of the sample, as illustrated in Figure 5.21. The photoassociation laser was illuminated on the sample for typically about 1.5 seconds with a power of approximately 40 mW. The remaining number of atoms in the lattice was monitored relative to the initially prepared atom quantity in the sample using fluorescence readouts. This approach allowed for the extraction of trap-loss spectra corresponding to the photoassociation lines.

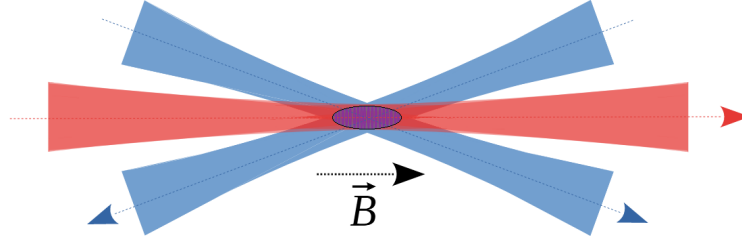


Figure 5.21: Top view of the optical lattice forming beams (blue) and the photoassociation beam (red) passing through the cesium sample confined in an optical lattice. The direction of the uniform magnetic field is shown with the black arrow.

The probed photoassociation spectra corresponding to two lowest-lying vibrational levels are shown in Figure 5.22 together with the upper vibrational level $\nu = 2$ and their energies are given in Table 5.3 with respect to the $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$ atomic transition.

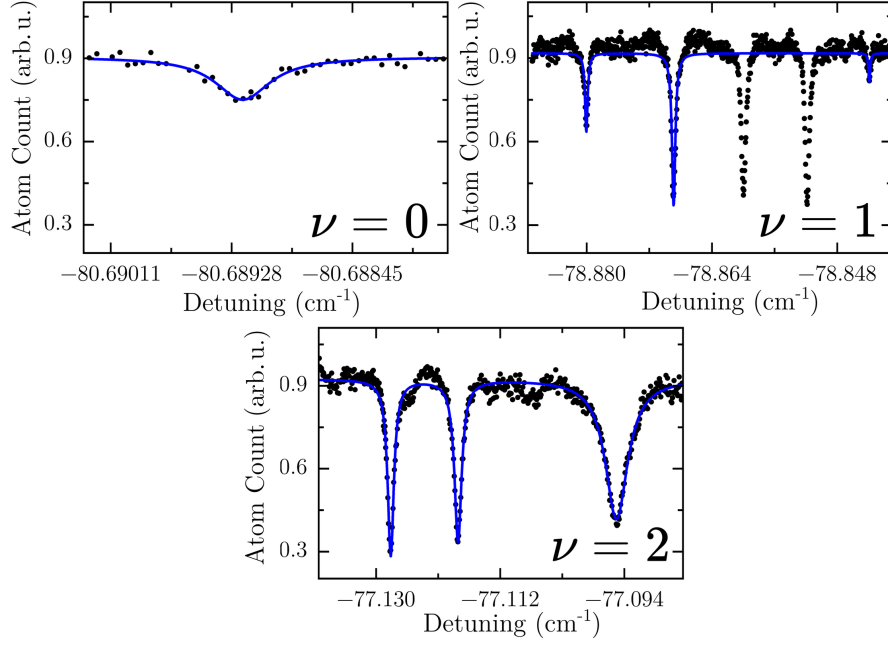


Figure 5.22: The measured photoassociation spectra for $\nu = 0$ ($J = 2$), $\nu = 1$ ($J = 0, 2, 4$) and $\nu = 2$ ($J = 0, 2, 4$) at the 19.7 G Feshbach resonance line of $F = 3$, $M_F = 3$ state. Detunings are referenced to the $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$ atomic transition. The blue lines are obtained by fitting a Lorentzian function to the observed peaks. Two unidentified photoassociation lines are present near the $\nu = 1$ rotational levels. The rotational numbering is determined based on the progression of rotational lines.

Table 5.3: The experimentally measured energies of the three lowest-lying levels ($\nu = 0$, $\nu = 1$, and $\nu = 2$) of the $\text{Cs}_2 0_g^-(P_{3/2})$ state, obtained at the 19.7 G Feshbach resonance (column Exp.). By compensating the Zeeman and AC stark shift, we determine the detuning of these levels in the absence of external effects (column Comp.). E_b is the binding energy of the corresponding rovibrational level measured with the energy scale being referenced to the $6s^2S_{1/2}(F = 4) \rightarrow 6p^2P_{3/2}(F' = 5)$ atomic transition.

(ν, J)	Exp. E_b (cm^{-1})	Comp. E_b (cm^{-1})
(0, 2)	-80.6892	-80.6909
(1, 2)	-78.8672	-78.8689
(2, 2)	-77.1164	-77.1181

In the rotationally resolved spectra, the lines corresponding to transitions to $J = 1$ and $J = 3$ levels are missing due to the polarization of the sample. The allowed values of J are $J = (l - 2)$, l , $(l + 2)$, where l characterizes a partial wave of the continuum state and $\vec{J}$ is the sum of $\vec{l}$ and of the total angular momentum of the electrons. For polarized bosonic particles, the symmetry forbids p -wave collisions [34, 207], which leads to the disappearance of transitions to $J = 1$ and

$J = 3$ levels. In our case, the efficiency of spin state polarization is considerably high. Thus, the intensities of the $J = 1$ and $J = 3$ rotational lines remained below the noise level, and, therefore, they were not observed. Additionally, using the same reasoning, the intensity of the $J = 2$ rotational line has been quasi-doubled. Since transitions to the $J = 4$ level are observed, a conclusion is made that the d -wave collisions also play a role in the photoassociation process. The photoassociation line intensities for the two lowest levels are very low, as expected. In fact, they could be detected only within a few hundred mG around the Feshbach resonance at 19.7 G (Figure 5.23).

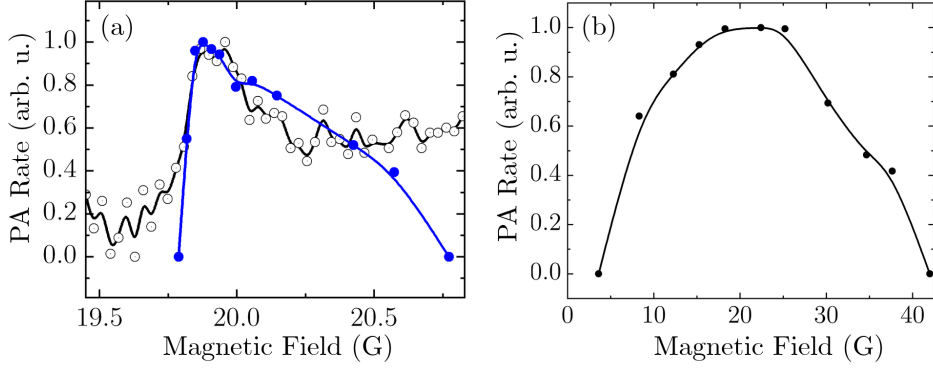


Figure 5.23: Intensities of photoassociation lines corresponding to two different vibrational levels ($\nu=0$ and $\nu=1$) of the $0_g^-(P_{3/2})$ state, when measured for various magnetic field values. (a) The photoassociation rate of $\nu = 0$ (blue) and the measured Feshbach resonance line (black) via the enhancement of the photoassociation rate of $1_g(P_{3/2})$ as a function of the applied external magnetic field. (b) The photoassociation rate of the $\nu = 1$ vibrational level as a function of the applied external magnetic field.

Since the $\nu = 1$ vibrational level corresponds to a much higher line intensity in comparison to $\nu = 0$, the enhancement of photoassociation with Feshbach resonances becomes less crucial for this vibrational level. Its rotational spectra were observable with our setup within a broader range of the applied external magnetic field. Additionally, the observability of the two lowest vibrational levels is anti-correlated with the photoassociation rate of the neighboring photoassociation transitions corresponding to the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. The experimental intricacies of these states were previously described in Section 5.5. Notably, a considerable increase in the photoassociation rate was observed in the case of vibrational levels of these states, coinciding with the Feshbach resonances at 47.9 G and 53.5 G. We have also measured the relation of the molecular formation rate versus the applied magnetic field of those neighboring levels of the $0_u^+(P_{3/2})$ state, which was, in fact, observed in Ref. [24] and erroneously labeled as $0_g^-(\nu=0$ and $\nu=1)$.

The enhancement of the photoassociation rate eventually broadens the spectral features corresponding to the neighboring $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states and blends the $0_g^-(P_{3/2})(\nu=0$ and $\nu=1)$ vibrational levels (Figure 5.24). The high molecular formation rate of the nearby states leads to a sudden drop in the density of our sample; thus, the $\nu=0$ and $\nu=1$ vibrational levels remain below the noise level. In fact, the squared values of Franck-Condon-Factors for the lowest three vibrational levels of the $0_g^-(P_{3/2})$ state were computed, mapping their dependence on applied external magnetic fields, which is described later in this section.

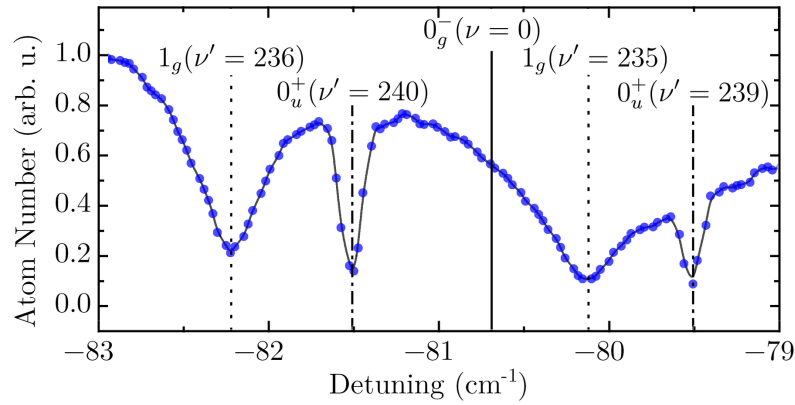


Figure 5.24: Trap-loss spectra of the 0_u^+ and 1_g states nearby the $0_g^-(\nu=0)$ vibrational level at the Feshbach resonance 47.9 G. Due to the significant enhancement of the neighboring PA lines, $0_g^-(\nu=0)$ vibrational level is not visible. For the states 0_u^+ and 1_g vibrational level labelling corresponds to $\nu' = [\nu_D - \nu]$.

As the experiments were conducted in the presence of external magnetic fields within an optical lattice, the resultant external factors were determined. Zeeman shift measurements were carried out by systematically scanning the laser frequency around the $\nu = 1, J = 2$ line while varying the magnetic field amplitude (Figure 5.25b). The resulting linear fit of the data yielded a slope of 2.11 ± 0.24 MHz/G, in good agreement with the theoretically predicted 2.1 MHz/G [248]:

$$\Delta\nu_{M_F} = 2 \times [(0.35 \text{ MHz/G}) \cdot M_F \cdot |\vec{B}|], \text{ where } M_F = 3. \quad (5.13)$$

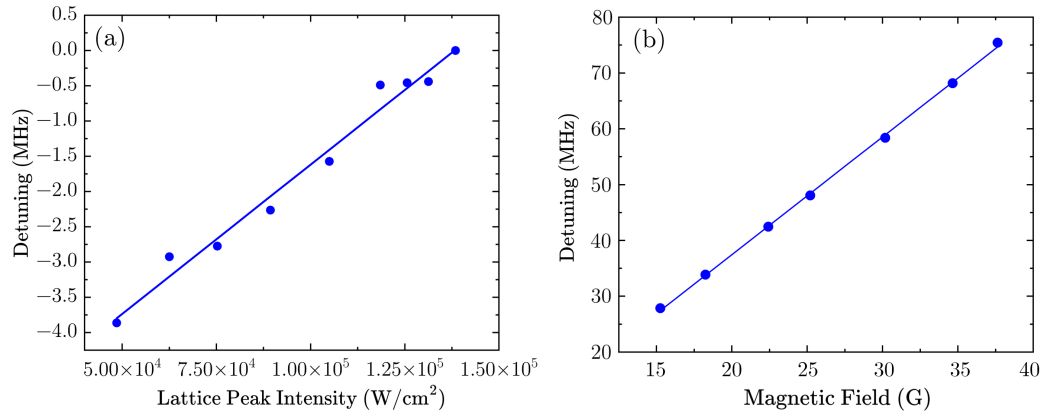


Figure 5.25: (a) Detuning of the $\nu = 1, J = 2$ rotational level as a function of the optical lattice peak intensity at constant magnetic field $B = 19.7$ G. (b) Detuning of the $\nu = 1, J = 2$ rotational level as a function of the applied external magnetic field.

Concurrently, AC Stark shift measurements were executed with a constant magnetic field, while varying the power of the beams generating the optical lattice. Through iterative measurements

of photoassociation spectra corresponding to the $\nu=1$, $J=2$ line, the effect of the AC Stark shift was determined (see Figure 5.25a). These determined values were then employed to subtract the external effects from the measured frequencies of the photoassociation rotational lines.

Following the above-mentioned measurements, a revision was undertaken to refine the determination of the potential energy curve for the $0_g^-(P_{3/2})$ state. The numerical calculations in this study were carried out using the analytical asymptotic approach as detailed in previous papers [21, 23]. The numerical computations were conducted in collaboration with our theoretical partner [249]. Additionally, it is worth noting that the analysis performed in this work closely follows the identical methodology outlined in Ref. [23].

In the applied model, the $0_g^-(P_{3/2})$ state was considered in Hund case (c) while being a result of the mixing of the $^3\Sigma_g^+(6S+6P)$ and $^3\Pi_g(6S+6P)$ electronic states which belong to the Hund's case (a). The mixing phenomenon can be described by a 2×2 Hamiltonian matrix [23]:

$$H = \begin{bmatrix} V^\Pi(R) - \Delta^{\Pi\Pi}(R) & \frac{\sqrt{2}M^2\epsilon}{9R^3} + \Delta^{\Pi\Sigma}(R) \\ \frac{\sqrt{2}M^2\epsilon}{9R^3} + \Delta^{\Pi\Sigma}(R) & V^\Sigma(R) \end{bmatrix}. \quad (5.14)$$

The external well of the $0_g^-(P_{3/2})$ potential has been described by asymptotic formulas, which involve the multipolar expansion of the electrostatic atom-atom interaction and the spin-orbit interaction. An asymptotic exchange interaction ($V_{exch}^{\Pi/\Sigma}$) is also considered as a perturbation. The equations defining the asymptotic potentials are given as;

$$V^\Pi(R) = -\frac{C_3^\Pi}{R^3} \left(1 + \frac{4\epsilon}{3}\right) - \frac{C_6^\Pi}{R^6} - \frac{C_8^\Pi}{R^8} + V_{exch}^\Pi, \quad (5.15)$$

$$V^\Sigma(R) = -\frac{C_3^\Sigma}{R^3} \left(1 + \frac{2\epsilon}{3}\right) - \frac{C_6^\Sigma}{R^6} - \frac{C_8^\Sigma}{R^8} + V_{exch}^\Sigma. \quad (5.16)$$

The equations above incorporate relativistic effects by a minor adjustment to the coefficients of the R^{-3} terms. This adjustment is characterized by the parameter ϵ , representing the ratio of squared transition moments ($M_{1/2}$ and $M_{3/2}$) associated with the relativistic $6P_{1/2}$ and $6P_{3/2}$ states. The equation describing the relationship between these parameters is

$$\frac{2\tau_{1/2}(\lambda_{3/2})^3}{2\tau_{3/2}(\lambda_{1/2})^3} = \frac{2}{(1+\epsilon)^2}. \quad (5.17)$$

Here, $\tau_{1/2}$ and $\tau_{3/2}$ are the atomic radiative lifetimes of corresponding states. The $(\lambda_{1/2}) = 11178.2682 \text{ cm}^{-1}$ and $(\lambda_{3/2}) = 11732.3071 \text{ cm}^{-1}$ are the wavenumbers corresponding to transitions from the atomic $6S_{1/2}$ level to $6P_{1/2}$ and $6P_{3/2}$, respectively [21]. The squared atomic transition dipole moment (M^2) is related to the coefficients $C_3^{\Sigma/\Pi}$:

$$C_3^\Pi = -\frac{C_3^\Sigma}{2} = -\frac{M^2}{3}. \quad (5.18)$$

The exchange interaction, denoted as $V_{exch}^{\Pi/\Sigma}$, is defined by the expression $V_{exch}^{\Pi/\Sigma} = -I_{no} - I_{ex}$, where I_{no} and I_{ex} represent the exchange integrals without and with excitation transfer, respectively. The exchange term depends on products of the amplitudes of the atomic asymptotic wave functions a_{6s} and a_{6p} . The mathematical formulations for the exchange integrals can be found in Ref. [21] and Ref. [112].

The spin-orbit coupling remains nearly constant across the entire range of the external well for the $0_g^-(P_{3/2})$ state. Consequently, the spin-orbit variation terms (A^{III} , $A^{\text{II}\Sigma}$) were set to an infinite value as done in the Ref. [23]. The spin-orbit interactions $\Delta^{\text{II}\Sigma}(R)$ and $\Delta^{\text{III}}(R)$ are given as:

$$\Delta^{\text{III}}(R) = \frac{\Delta E_{fs}}{3} \tanh(A^{\text{III}}R), \quad (5.19)$$

$$\Delta^{\text{II}\Sigma}(R) = \frac{\sqrt{2}\Delta E_{fs}}{3} \tanh(A^{\text{II}\Sigma}R), \quad (5.20)$$

where, $\Delta E_{fs} = 554.039 \text{ cm}^{-1}$ is the cesium $6P_{3/2}$ fine structure splitting [45]. Diagonalization of the 2×2 matrix in Equation 5.14 yields an analytical expression for the potential curve of the 0_g^- external well. Vibrational energies and wave functions are then calculated using the standard Numerov integration method. Following the approach outlined in Ref. [23], a minimization procedure (fitting of variables to minimize rms value) based on the generalized simulated annealing (GSA) method [250] was employed for fitting the experimental values. This method, renowned for its ease in handling nonlinear fits, was utilized to characterize the long-range behavior of molecular potentials [250, 251]. The root-mean-square for each rovibrational level is determined as the following, where N_p is the count of parameters that are adjusted to minimize the root-mean-square difference between the N calculated and N experimental energies:

$$\text{rms} = \sqrt{\frac{\sum_{i=1,N} (E_{\text{exp}} - E_{\text{cal}})^2}{N - N_p}}. \quad (5.21)$$

In the fitting procedure via GSA, we have used the energies of 71 previously measured [252] and the two newly measured (this work) levels to determine the squared atomic transition moment M^2 , the relativistic parameter ϵ , multipole expansion coefficients, C_3^Σ , C_3^Π , C_6^Σ , C_6^Π , C_8^Σ , C_8^Π , and the exchange amplitude $a_{6s}a_{6p}$.

In the context of our study, the equilibrium point is determined to be at $12.41 \text{ \AA}$ ($23.46a_0$) with a corresponding potential energy value of -81.735 cm^{-1} . In comparison, the previously reported value at $12.41 \text{ \AA}$ yields -81.644 cm^{-1} [23], demonstrating a close agreement. We introduced parameters κ_R and κ_D to serve as indicators of sensitivity regarding the minimum position (R_e) and potential depth (D_e) with respect to the fitted parameters of the model. The sensitivity values are calculated using the following relationships:

$$\frac{dR_e}{R_e} = \kappa_R \frac{d\xi}{\xi}, \quad (5.22)$$

$$\frac{dD_e}{D_e} = \kappa_D \frac{d\xi}{\xi}, \quad (5.23)$$

where, ξ represents the respective variable from [Table 5.4](#).

Table 5.4: The parameters κ_R and κ_D are evaluated for various variables of the external well within the $0_g^-(P_{3/2})$ state. These values are determined by holding the remaining variables constant and identifying the location of the potential curve's minima along with its corresponding depth.

Variable	κ_R	κ_D
ϵ $[10^{-3}]$	0.0033	-0.013
$M^2[10^5 \text{Å}^3 \text{cm}^{-1}]$	0.64	-0.73
$C_6^\Sigma[10^7 \text{Å}^6 \text{cm}^{-1}]$	-0.008	0.064
$C_8^\Sigma[10^9 \text{Å}^8 \text{cm}^{-1}]$	-0.024	0.083
$C_6^\Pi[10^7 \text{Å}^6 \text{cm}^{-1}]$	-0.088	0.15
$C_8^\Pi[10^9 \text{Å}^8 \text{cm}^{-1}]$	-0.044	0.06
$a_{6s}a_{6p}$	0.028	-0.029

The results indicate a potential mislabeling of rotational quantum numbers for the observed vibrational levels ($\nu = 0$ and $\nu = 1$). The measured $J = 2$ of the $\nu = 0$ level suggests it could actually be $J = 4$, while the measured $J = 0, 2, 4$ in the $\nu = 1$ level might correspond to $J = 4, 5, 6$ respectively. In fact, all the numerical fitting methods have given lower root-mean-squares when done with these suggested numbering of rotational numbering. Thus, the results of the fitting process presented in this thesis were given together with the suggested values (see [Figure 5.26](#)). These levels, situated at the bottom of the external well of the $0_g^-(P_{3/2})$ state, exhibit remarkable sensitivity to the potential minimum and its associated internuclear distance.

When the assignment of the rotational numbering is considered with the rotational constant B_{rot} , where $E_{rot} = B_{rot}[J(J+1) - \Omega^2]$, it favors the numbering given as $J=0, 2, 4$ in the $\nu = 1$ vibrational level. In fact, these lines correspond to s -wave and d -wave collisions since p -wave collisions are suppressed as described in [Section 5.3](#). The emergence of the weak $J = 4$ component may stem from d -wave collisions, characterized by a barrier height of approximately $200 \mu\text{K}$, involving two rapidly moving atoms within the thermal distributions [\[13\]](#). Considering the initial temperature of our sample to be $12 \mu\text{K}$, it's improbable for higher-order collision waves, such as f -wave and g -wave, to influence the photoassociation process. Consequently, we anticipate that the f -wave and g -wave collisions do not play a role in the photoassociation process, disallowing the observation of $J = 5$ and $J = 6$ rotational energy levels. Additionally, due to the quasi-doubling of the $J = 2$ rotational energy level, we favor labeling the rotational levels, including this particular rotational level, where its line intensity is expected to be highest among other rotational levels. For this reason, the only probed rotational level in the $\nu = 0$ is labeled as $J = 2$.

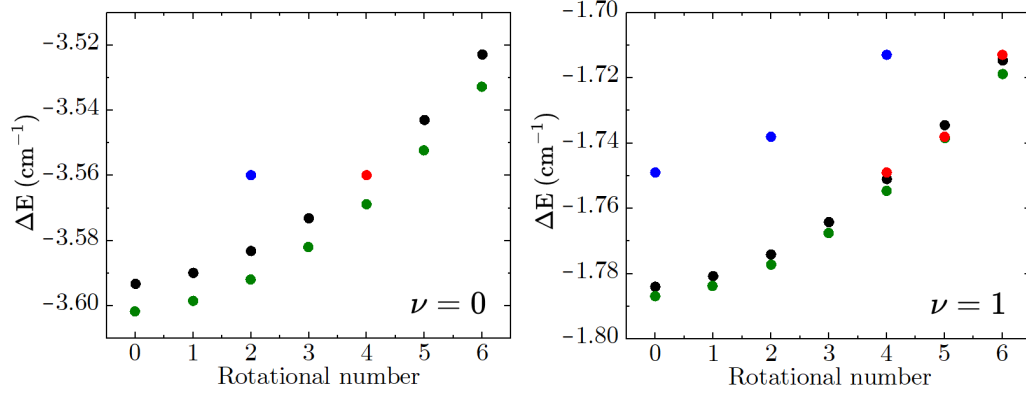


Figure 5.26: The rotational energy levels for $\nu = 0$ (left) and $\nu = 1$ (right) in the external well of $0_g^-(P_{3/2})$ state when numerically determined with the suggested rotational numbering yielding minimum residuals shown in reference to $\Delta E(\nu, J) = E(\nu, J) - E(2, 0)$. The black data points represent the rotational energies calculated from the numerical fitting conducted in this work. The green data points represent the rotational energies determined with the parameters reported in Ref. [23]. The blue data points illustrate the rotational levels measured in this study, while the red data points represent the suggested rotational numbering that results in reduced error.

Table 5.5: The table presented here showcases the outcomes of our optimal fitting, along with results from Ref. [21, 23, 112, 243]. Notably, the values of the parameters that have been kept fixed are indicated within curved parentheses.

Variable	Theory	Ref. [21]	Ref. [23]	This work
$\epsilon[10^{-3}]$	-	4.69	(4.81)	(4.81)
$M^2[10^5 \text{Å}^3 \text{cm}^{-1}]$	10.22[243]	9.798	9.806	9.808
$C_6^\Sigma[10^7 \text{Å}^6 \text{cm}^{-1}]$	8.381[243]	8.506	8.788	7.462
$C_6^\Pi[10^7 \text{Å}^6 \text{cm}^{-1}]$	5.701[243]	5.689	5.869	6.215
$C_8^\Sigma[10^9 \text{Å}^8 \text{cm}^{-1}]$	6.802[243]	7.715	17.525	16.34
$C_8^\Pi[10^9 \text{Å}^8 \text{cm}^{-1}]$	3.045[243]	3.18	3.522	3.820
$a_{6s}a_{6p}$	0.054790[112]	0.03843	0.04512	0.0453

Even in the context of the best fitting (results are presented in Table 5.5), these parameters are significantly influenced by 71 previously obtained data points from other studies, each carrying an uncertainty of approximately ± 60 MHz. This uncertainty significantly influences the accuracy of predictions for the rotational levels in the two lowest vibrational levels. The inherent variability in the prior data introduces challenges in accurately determining rotational quantum numbers. Consequently, this necessitates the repetition of previously conducted photoassociation experiments with higher precision to accurately establish the rotational numbering. The residuals depicted in Figure 5.27 showcase a comparison between the rotational energy levels obtained from the previously reported 202 rotational energy levels [22] and the four rotational energy levels probed in this study, in relation to the fitting conducted using parameters from Ref. [23].

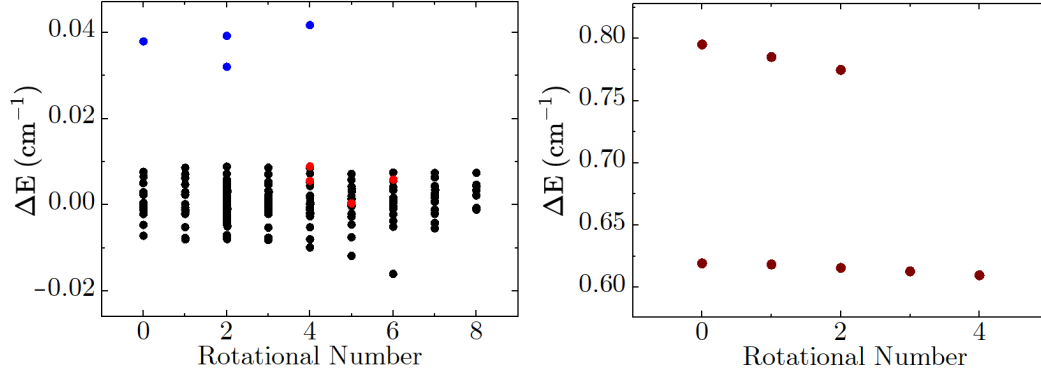


Figure 5.27: (a) Residuals for all 202 rotational energy levels measured in the photoassociation experiment of Ref. [22] (in black) were digitized from Ref. [23], along with the residues of four levels obtained in this work (in blue), and suggested rotational numbers from our numerical fit (in red). (b) Residuals of the measured levels reported by Ref. [24] are determined using the potential energy curves presented in Ref. [23].

The residuals highlight discrepancies between the fitted values and the actual measurements. Notably, the uncertainties of ± 60 MHz introduce higher-order errors during the fitting process, surpassing the precision of previous measurements. It is worth noting that the previously reported experimental works [24] demonstrate significantly larger residuals values compared to the numerically expected values. This discrepancy arose due to their mislabeling as the lowest lying vibrational levels of $0_g^-(P_{3/2})$ instead of $0_u^+(P_{3/2})$. Despite the potential energy curve and the fitting process being more sensitive to the vibrational levels at the bottom of the external well of the $0_g^-(P_{3/2})$ state, the reported parameters resulting from the fit done by Zhang et al. [24] were similar to those reported in Ref. [23]. This consistency of the parameters used in the fitting process is unexpected, given the existence of such high residuals for the lowest two vibrational levels.

From the analysis made, one can determine the atomic lifetime values from the value of M^2 :

$$\tau_{3/2} = \frac{9\hbar}{4M^2} \left(\frac{\lambda_{3/2}}{2\pi} \right)^3, \quad (5.24)$$

$$\tau_{1/2} = \tau_{3/2} \left(\frac{\lambda_{1/2}}{\lambda_{3/2}} \right)^3 \frac{1}{(1 + \epsilon)^2}. \quad (5.25)$$

By using the parameters obtained from the fit and presented in Table 5.5, the corresponding atomic lifetimes are determined as $\tau_{3/2} = 29.17 \pm 0.29$ ns and $\tau_{1/2} = 33.4 \pm 0.35$ ns. Table 5.6 and Figure 5.28 show the comparison of the determined radiative lifetimes to previously reported values.

The values of the van der Waals coefficient C_6 for the ground state, along with the atomic lifetimes of $6p^2P_{1/2}$ and $6p^2P_{3/2}$, remain in agreement with existing data, though yet again with moderate precision. It is important to note that our results, while not providing conclusive differentiation

Table 5.6: Radiative lifetimes of the cesium atomic $6p^2P_{3/2}$ and $6p^2P_{1/2}$ states are compared with previously reported values, along with the values of the relativistic parameter ϵ in which if in brackets are kept as constant during the numerical fitting procedure.

	$\tau_{3/2}(\text{ns})$	$\tau_{1/2}(\text{ns})$	$\epsilon(10^{-3})$
This Work	30.42(0.30)	34.83(0.36)	(4.81)
Ref. [3]	30.57(0.07)	35.07(0.10)	3.92(2.60)
Ref. [4]	30.41(0.10)	34.75(0.07)	5.88(2.60)
Ref. [21]	30.462(0.003)	34.89(0.02)	4.69(0.03)
Ref. [23]	30.41(0.30)	34.82(0.36)	(4.81)
Ref. [253]	30.39(0.06)	34.80(0.07)	4.81(2.10)

among various atomic measurements due to limited precision, exhibit strong consistency with the intensity patterns observed in multiple experimental spectra. This consistency serves as a compelling argument in favor of the reliability of our analysis, especially when the applied external magnetic fields are included in the measurements.

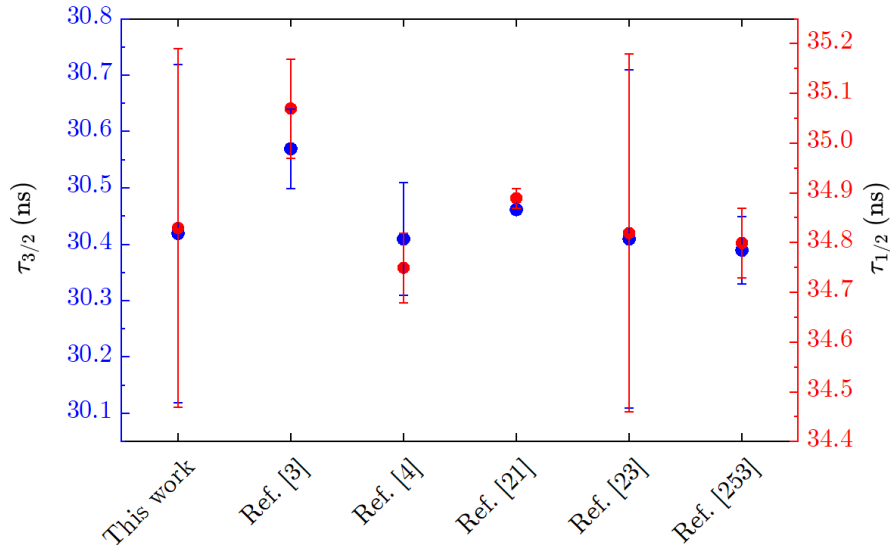


Figure 5.28: Numerically determined radiative lifetimes of the cesium atomic $6p^2P_{3/2}$ and $6p^2P_{1/2}$ states in comparison with previously reported values [3, 4, 21, 23, 253].

The analysis of the photoassociation line intensities is done by determining the Frack-Condon factors in relation to the overlap of the excited state and ground state wavefunctions. For each applied magnetic field value, the ground state wavefunction has been recalculated, and the resulting overlapping has been reevaluated. The results demonstrated a correlation with our experimental results (refer to Figure 5.29).

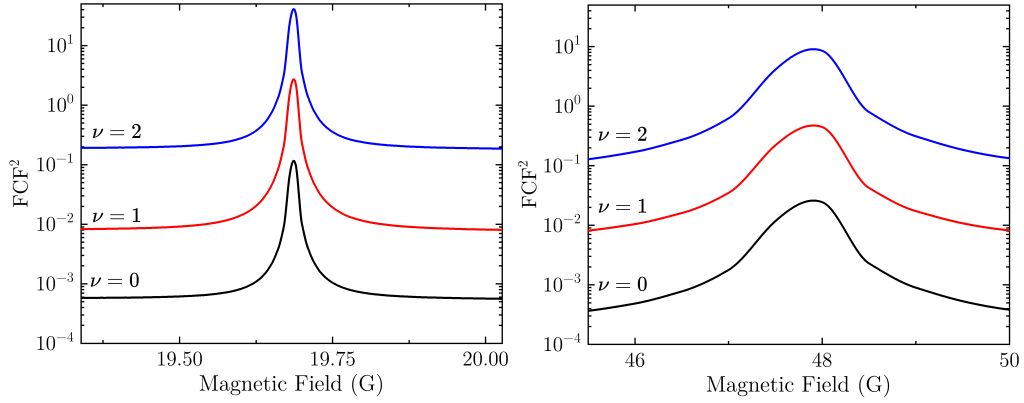


Figure 5.29: Squared values of Frack-Condon-Factors corresponding to transitions to the three lowest vibrational levels of $0_g^-(P_{3/2})$ state given as a function of applied external magnetic fields. The data shows significant enhancement of photoassociation rates upon crossing Feshbach resonances of cesium atoms in the energy level $F = 3, M_F = 3$.

Our experimental setup has demonstrated its capabilities by successfully measuring previously unobserved vibrational levels with high precision. Moreover, revisiting previously measured spectra has proven valuable for determining atomic constants such as radiative lifetime. Typically, photoassociation spectroscopy experiments are not frequently repeated by different research groups unless a particular vibrational level exhibits notable properties, such as a high efficiency of the STIRAP process for potential applications. Consequently, technological advancements in this field are often underutilized for such experiments. In principle, the constructed setup is not only beneficial for future experiments but, in fact, can be used for the analysis of previously measured photoassociation spectra to minimize the uncertainties in the determination of such atomic constants.

Chapter 6

Conclusion and Outlook

In conclusion, this thesis presents an investigation of the photoassociation of Cs_2 molecules, focusing on the external well of the 0_g^- long-range state dissociating towards the $6S_{1/2} + 6P_{3/2}$ asymptote. A notable outcome of this work is the first observation and accurate measurement of energies of the two lowest vibrational levels situated in the $0_g^-(P_{3/2})$ external well. This has resulted in the resolution of a discrepancy in the literature and the confirmation of a theory that has been in existence for over a decade. We also provide unambiguous evidence that previously reported experimental works that claimed the priority in observing these new levels, in fact, observed levels belonging to other molecular states. The experimental apparatus designed for this purpose not only successfully probed the lowest lying previously unobserved vibrational levels of the $0_g^-(P_{3/2})$ state, but also demonstrated its versatility by extending measurements to these other molecular states – $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ – down to a detuning of -300 cm^{-1} below the dissociation limit. This accomplishment was made possible through the progressive development of our experimental apparatus by the implementation of advanced techniques in photoassociation experiments, spanning improvements in cooling methods of cesium atoms, experimental control schemes, and imaging sequences.

To summarize these developments, apart from the construction of the base components of our sample preparation apparatus, the progress in our experimental setup for photoassociation experiments began by addressing challenges with the assembled titanium:sapphire lasers, ensuring consistent operation by making proper adjustments on their hardware. The establishment of an optical system for light distribution and laser frequency measurement was designed and built for these lasers. Their outputs were referenced with an optical frequency comb for high-precision photoassociation spectroscopy, marking significant progress in the reliability of our measurements.

During the early phases of our photoassociation experiments within the KCs single chamber, our capabilities were initially confined to conducting experiments solely within the confines of the MOT. Subsequently, experimental progress in loading our ultracold sample into an optical dipole trap resulted in colder and denser samples, further enhancing the capabilities of the photoassociation setup. Additionally, it enabled precise control over collisional properties through the application of an external magnetic field. The availability of spin polarization offered improved control over the initial state of the atoms, revealing previously reported and observed Fesh-

bach resonances that remained unseen through the experiments conducted in our setup. The experimental focus on photoassociation spectroscopy then shifted to the implementation of the enhancement method ‘Feshbach optimized photoassociation’, specifically to be able to probe the lowest two vibrational levels of the $0_g^-(P_{3/2})$ state in which both vibrational levels demonstrate remarkably low transition probability. After incorporating the aforementioned advancements into our setup, we were finally able to probe these vibrational levels and extend the observed range of $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states.

Subsequent to these novel observations, our focus shifted towards a numerical analysis of the experimentally measured rovibrational data. The analysis of the potential energy curve of the $0_g^-(P_{3/2})$ state yielded results in agreement with the previously developed theoretical postulates. The analysis has also uncovered potential discrepancies in the rotational level numbering compared to our initial assumptions. It has been established that, for further refinement of the precision in our analysis, it is imperative to improve the accuracy of previously recorded data measurements to enable more thorough assessments. Furthermore, our analysis of Feshbach-optimized photoassociation yielded expected results, particularly when determining the photoassociation rate of the lowest vibrational levels with changes in the ground state wavefunction due to the presence of an applied external magnetic field. At present, the project is ongoing, as we require measurement of several vibrational levels near the bottom of the potential with high resolution to assess the theoretical analysis further. It should be noted that the theoretical analysis has demanded more time than initially anticipated, but we anticipate concluding this aspect shortly. Subsequently, the publication of a research paper stemming from this work will follow once our analysis is complete.

The data obtained in the experiments extending the measurements of the states $1_g(P_{3/2})$ and $0_u^+(P_{3/2})$ were not rotationally resolved and exhibited spectral broadenings. The broadening of these transitions is attributed to predissociation phenomena associated with the avoided crossing of $0_g^+(P_{1/2})$ for the $1_g(P_{3/2})$ state and to interaction with the $0_u^+(P_{1/2})$ state for the $0_u^+(P_{3/2})$ state. The data obtained from these measurements underwent analysis using several theoretical models. However, models for potential energy curves of the relevant molecular states did not yield satisfactory results due to the lack of experimental data in the intermediate internuclear region between the short and long range. The upper high-lying vibrational energies were probed in the long-range region, where the theoretical models were intended to work the best. Numerical fitting applied to high-lying vibrational levels ($\nu \leq 255$) produced consistent results. However, for the spectra reached below -120 cm^{-1} , the probed states were considered to be in the intermediate range, leading to a sudden increase in error in the numerical fitting process. Nevertheless, to come, the vibrational numbering assigned as a result of our analysis of these levels appears to be unambiguous. In addition, it is worth mentioning that a potential publication opportunity exists for the measurements extending to the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states. Specifically, an experimental approach to measuring all observable vibrational levels of the $0_u^+(P_{3/2})$ and $1_g(P_{3/2})$ states could be pursued following theoretical work to improve molecular potential modeling suitable for the range transitioning from long-range to intermediate-range. However, due to time constraints and the primary focus of this thesis is the $0_g^-(P_{3/2})$ state, we have not pursued this possible approach thus far.

Looking ahead, the photoassociation setup developed in this work holds great potential for future investigations, especially in measuring photoassociation levels of KCs molecules in the near future within the same experimental apparatus. The advancements made in this thesis contribute not only to the comprehension of Cs_2 molecular states but also establish the foundation

for further experiments in our research group, specifically the formation of ground-state KCs molecules. Additionally, a publication discussing the collisional properties via the simultaneous magneto-optical trapping of cesium and potassium isotopes is currently in progress as a part of the contributions made to our apparatus through this thesis. The constructed apparatus also demonstrates the potential for integration into our group's newly launched research project, which is centered on studying ultracold KAg and CsAg molecules.

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