#### VRIJE UNIVERSITEIT

## High-Precision Spectroscopy of Forbidden Transitions in Ultracold <sup>4</sup>He and <sup>3</sup>He

ACADEMISCH PROEFSCHRIFT

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voor mijn familie en vrienden

Science is an ongoing process. It never ends. There is no single ultimate truth to be achieved, after which all the scientists can retire. Carl Sagan, Cosmos This thesis was approved by the members of the reviewing committee:

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The cover shows the observed absorption line shapes of a narrow optical transition in a degenerate Fermi gas (top) and a Bose-Einstein condensate (bottom) of metastable helium. The large difference is caused by the fundamental difference in quantum statistics between fermions and bosons.

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## CHAPTER 1

## Introduction

#### 1.1 Prologue

Science is in perpetual motion, and changes in our fundamental understanding of the workings of the physical universe are inevitable as we are presented with new evidence through observations. Of all the methods an experimental physicist has in his toolbox, spectroscopy is the most accurate tool available to observe the universe. Applied to a suitable test object, whose measurable quantities can be calculated to high precision, physical theories can be tested to extreme limits within the confines of a (relatively small-scale) laboratory setup.

The test object in this thesis is the helium atom, which is a threebody system consisting of a nucleus and two electrons. This makes it simple enough that high precision calculations can be performed for this system, yet it contains sufficient complexity to be challenging on both the theoretical and experimental front. My work in this thesis focuses on high-precision spectroscopy in the helium atom, and in this introduction I provide an overview of the many different facets of fundamental physics where the helium atom plays an important role. But first I will discuss the essential role that spectroscopy played in the 'quantum-revolution' of the 20th century, and how it still does to this date.

#### **1.2** A revolution in physics

A little over a century ago there was general consensus that mankind's understanding of physics was nearly complete. With the grand successes of Maxwell's laws of electromagnetism, Newton's laws of gravity and motion, and the framework of thermodynamics, there were few gaps to be filled to complete our view of the workings of the universe. This is immortalized by Lord Kelvin's - the worlds' foremost natural philosopher at the time - speech in 1900 in which he states that physics is understood apart from two 'clouds' [1]. The first cloud was caused by the famous experiment by Michelson and Morley which seemed to disprove the 'luminiferous ether' theory as no translational variation in the speed of light could be observed. The second cloud was related to the Maxwell-Boltzmann theory of equipartition of energy, which could not explain the specific heat properties of matter based on a continuum distribution of energy states.

Within five years, four papers were published by Albert Einstein to address these clouds and they sparked a revolution [2–5]. In the next decades our understanding of the physical world would transform from a deterministic view to the exciting yet astonishing world of quantum and relativistic mechanics, and even relativistic quantum mechanics. This revolution is now associated with names like Bethe, Bohr, Born, de Broglie, Dirac, Ehrenfest, Fermi, Feynman, Heisenberg, Lamb, Pauli, Planck, Schrödinger, and Sommerfeld, and I am definitely doing many other physicists wrong by reducing the contributions to this single list. Some of these names will reappear multiple times in this thesis.

The initial test model for the theoretical developments was the hydrogen atom. This is understandable from a theoretical point of view, as the two-body system (one electron and one proton) allowed exact analytical solutions of the electronic level energies. As the energy difference between energy levels could be measured using spectroscopy, the early success of quantum mechanics was the explanation of the hydrogen spectrum using the model proposed by Bohr and the resulting derivation of the empirical Rydberg constant in terms of fundamental constants [6]. This paved the road for spectroscopy as the main experimental force behind further refinement and development of the theory. Although very successful, there already were some problems with the theory proposed by Bohr as it could not explain the presence of so-called 'doublet' lines that had been observed in hydrogen spectra already in 1887 [7]. There soon appeared an explanation by Sommerfeld by introducing elliptical Keplerian orbitals for the electrons [8], but the real breakthrough was made by Dirac, who proposed a relativistic quantum mechanical description of the electron [9]. In this model the observed fine structure in the hydrogen energy levels could be explained by spin-orbit coupling, and the degenerate 2S and 2P levels in hydrogen would actually be split into the 2  ${}^{2}S_{1/2}$ , 2  ${}^{2}P_{1/2}$ , and 2  ${}^{2}P_{3/2}$  levels where only the 2  ${}^{2}S_{1/2}$  and 2  ${}^{2}P_{1/2}$  levels would remain degenerate.

Within 30 years since the speech by Lord Kelvin, our understanding of physics was completely revolutionized both in the realm of the extremely small and the extremely fast. And yet even the model proposed by Dirac would soon appear incomplete due to advances in the spectroscopic techniques. In 1947 Lamb and Retherford finished an experiment from which they concluded that the hydrogen 2  ${}^{2}S_{1/2}$  and 2  ${}^{2}P_{1/2}$  levels were not degenerate but actually split by 0.033 cm<sup>-1</sup> (~ 1 GHz) [10, 11]. The explanation of this so-called 'Lamb shift', given by Bethe [12], required a new mathematical framework where quantum mechanics and electrodynamics were incorporated into a single operating model: quantum electrodynamics (QED).

As if quantum mechanics was not weird enough, QED tells us that the vacuum is not empty. Given sufficiently short time, particles can 'pop up' in a vacuum and disappear a short time later without violating any conservation laws. However, these so-called 'virtual' particles can still interact with other particles, and lead to shifts in the electronic level energies of atoms. The observation of the Lamb shift accelerated the development of QED as laid out by Tomonaga, Schwinger, and Feynmann. One of the earliest overviews of the theory - authored by Dyson would be published a mere two years after the observation of the Lamb shift [13]. By this time the expected value for any observable parameter could be calculated in QED theory as a series expansion of contributing processes as function of powers of the fine structure constant  $\alpha$ , where each higher order involves more complex processes. As there is no a *priori* reason to accept that the series expansion converges [14], higherorder terms can still significantly contribute and require evaluation and continuous comparison with increasingly accurate measurements.

Apart from the Lamb shift, another puzzling discrepancy in the 1940's was the observation that the magnetic moment of an electron was different from the expected value [15–17]. Within a few years this deviation was shown to be in agreement with new experiments and fourth-order QED calculations [18, 19]. Together with the explanation of the Lamb shift, the validity of QED was established and continued to provide agreement between theory and experiment for decades to come. By 2012, 65 years after the first results, QED calculations up to tenth order contributions (which involves 12 672 different QED processes (Feynman diagrams) for this order alone) can predict the anomalous magnetic moment with over 9-digit precision [20]. Even more staggering is the fact that this value is in good agreement with experiment [21]. There are very few theories in physics that have survived so many orders of magnitude improvement in experimental accuracy and are still able to correctly predict the outcome.

#### 1.3 Pushing QED to the limit

Spectroscopy has been an important experimental tool to help develop the rapidly changing and expanding world of quantum and relativistic mechanics and finally quantum electrodynamics. The robustness of QED also gives rise to a curiosity; how far can we go before the theory will break down? And will we encounter new physics?

In a quest for pushing QED to its limit to answer these questions, spectroscopic techniques have advanced to the point where experimental results can be much more accurate than the QED predictions, simply because the input parameters of the theory (i.e. fundamental constants) are known with insufficient accuracy. We are now in an interesting regime where we might want to reverse the former question: if QED is correct and our experiments are correct, what could we learn about the fundamental constants? I will discuss both scientific lines of investigation here, and focus on applications with helium.

#### Direct test of QED

Apart from hydrogen as a testing ground for the development of quantum mechanics and QED, the helium atom quickly gained interest as well. Exact solutions are unavailable as the atom is a three-body system, and perturbative approaches are required. As soon as in the early 1950's the ionization energies of the ground  $(1 \ {}^{1}S_{0})$  and first excited  $(2 \ {}^{3}S_{1})$  states of helium were investigated both experimentally and theoretically (see, e.g. Refs. [22–24]). For light atomic systems such as the hydrogen or helium atom, the relativistic and QED effects can be calculated as small perturbations of the nonrelativistic eigenenergies of the atomic system and expressed as an expansion series in powers of the fine structure constant  $\alpha$  as [25, 26]

$$E(\alpha) = \sum_{i} m_e \alpha^i \mathcal{E}^{(i)}, \qquad (1.1)$$

where  $m_e$  is the electron mass.  $\mathcal{E}^{(i)}$  can again be expanded as a power series in terms of the electron-to-nucleus mass ratio  $m_e/M$ 

$$\mathcal{E}^{(i)} = \mathcal{E}^{(i)}_{\infty} + \mathcal{E}^{(i)}_{M} + \mathcal{E}^{(i)}_{M^2} + \dots$$
(1.2)

The series expansion looks straightforward, but each term involves its own effective operator including nonanalytic terms. For neutral helium the perturbations can nowadays be calculated to 6<sup>th</sup> order [26, 27] with very recent progress to 7<sup>th</sup> order [28], at which the low-lying energy levels (with principal quantum number n = 1, 2) can be calculated with accuracies ranging from 36 MHz for n = 1 to  $\sim 1$  MHz for n = 2 [26]. The QED contributions are largest for these low-lying states, and decrease for states with increasing n and angular momentum L. As the higher-lying state energies can be calculated with much better precision than the low-lying states, they can be used as 'anchors' for testing the accuracy of the QED calculations of the low-lying states by measuring transition frequencies between low-lying and high-lying states. An example is the  $3^{3}D_{1}$  state, which is known to 20 kHz accuracy and used to determine the ionization energy of the 2  ${}^{3}S_{1}$  state to 60 kHz accuracy [30, 32], much better than the theoretical accuracy of 2.6 MHz [26]. There is a large body of work involving many measured transitions to test the theory, of which Refs. [29, 31–44] represent the contributions of the last 25 years. In parallel the calculations improved significantly after a breakthrough in the variational calculation techniques by Drake

State	Experiment [MHz]	ExpTh. [MHz]	Ref.
$1  {}^{1}S_{0}$	$5945204212$ $\pm 6$	$38 \pm 36$	[29, 30]
$2 \ {}^1S_0$	$960332040.71\pm0.06$	$2.7 \hspace{0.2cm} \pm 1.9 \hspace{0.2cm}$	[31]
$2 {}^{3}S_{1}$	$1152842742.97\pm0.06$	$1.5 \pm 2.6$	[30, 32]
$2 {}^{1}P_{1}$	$814709147.9\ \pm 0.2$	$1.5 \pm 0.4$	[33]
	$814~709148.1~\pm 0.3$	$1.7 ext{ }\pm 0.5 ext{ }$	[30, 34]
	$814709148.5\ \pm 0.5$	$2.0 \hspace{0.2cm} \pm 0.7 \hspace{0.2cm}$	[35]
$2 \ ^{3}P$	$876106247.21\pm0.06$	$1.2 \hspace{0.2cm} \pm \hspace{0.2cm} 0.9 \hspace{0.2cm}$	[36]

**Table 1.1** – The most accurate experimental values of the ionization energies of the n = 1, 2 levels in <sup>4</sup>He. The ionization energies in italics are determined by combining measurements with accurate calculations of higher states [30], and the other ionization energies are determined using the 2 <sup>3</sup>S<sub>1</sub> state as an anchor and combining measured transition frequencies (which are also shown in Figure 1.1). The experimental results are compared to the most accurate QED calculations available [26]. The 2 <sup>3</sup>P centroid energy is defined as  $\frac{1}{9}[E(2 \ {}^{3}P_{0}) + 3E(2 \ {}^{3}P_{1}) + 5E(2 \ {}^{3}P_{2})]$ .

in 1987 [45, 46], which allowed the nonrelativistic eigenvalues to be calculated to virtually any numerical accuracy. The theoretical developments in the calculation of relativistic and QED effects, in particular by Pachucki, would result in a more than tenfold improvement in the accuracy in the next decades [26–28, 30, 47–57].

As there has been no general overview of theory and experiment of the low-lying levels in literature since 2008 [57], the current most accurate experimental determinations of the ionization energies of the n = 1, 2states in <sup>4</sup>He are listed in Table 1.1 and compared to theory, and Figure 1.1 shows the measured transitions on which these energies are based. For the determination of the ground state energy of helium a comprehensive historic overview is given in Ref. [58], and the most accurate determination to date is based on a direct measurement of the 1  ${}^{1}S_{0} - 5 {}^{1}P_{1}$ transition at 51.5 nm [29] combined with theory [30]. The resulting 6 MHz accuracy in the experiment is much better than the most accurate calculation to date (36 MHz accuracy) [26], and both results are in agreement. For the 2  ${}^{3}S_{1}$  ionization energy the most accurate determi-



**Figure 1.1** – Low-lying energy levels in <sup>4</sup>He that are used to test QED. The lines indicate the transitions and their respective wavelengths used to determine the ionization energies (see Table 1.1) experimentally.

nation is based on the measurement of the 2  ${}^{3}S_{1} - 3 {}^{3}D_{1}$  two-photon transition [32] combined with theory for the 3  ${}^{3}D_{1}$  state [30] to 60 kHz accuracy. The most precise calculation with an accuracy of 2.6 MHz [26] is in agreement with this result. The 2  ${}^{3}S_{1}$  state is of special interest as it has a lifetime of  $7.8 \times 10^{3}$  s [59] and is used as a long-lived metastable state (generally denoted as He<sup>\*</sup>) from which optical spectroscopy can be performed at wavelengths which are much easier to generate than the short wavelengths required for ground-state spectroscopy (see Figure 1.1). The He<sup>\*</sup> state plays the central role in this thesis.

Until 2011 the determination of the ionization energies solely depended on the measurements in the separate triplet or singlet Rydberg series. The measurement of the  $2 {}^{3}S_{1} - 2 {}^{1}S_{0}$  and  $2 {}^{3}S_{1} - 2 {}^{1}P_{1}$  intercombination lines in our group [31, 35] opened the path for direct comparison of ionization energies between the different series, and a direct link between the accurate 2  ${}^{3}S_{1}$  ionization energy and the 2  ${}^{1}S_{0}$  and 2  ${}^{1}P_{1}$  states [31, 33, 35]. This network of transitions, as shown in Figure 1.1, provides a self-consistent set of measurements to test QED. As can be seen in Table 1.1, the largest discrepancy (~  $3\sigma$ ) between theory and experiment is presently in the 2  ${}^{1}P_{1}$  state which will be further discussed in Chapter 3.

#### 1.4 Determining fundamental constants

The advances in the theory in the last decades are big, and the experimental accuracies have improved as well. Significant theoretical improvements are now required in order to test QED in the helium atom further. As we currently find general agreement between theory and experiment, one can wonder if we cannot use the experimental values to extract information about the fundamental parameters that are at the base of the theory.

This approach has been used in other systems. For example, the earlier mentioned anomalous magnetic moment of the electron presents a testing ground where both QED and experiment reach such high precision and agreement that the fine structure constant can be determined with  $2.5 \times 10^{-10}$  relative accuracy [20, 21]. Helium can be used for similar tests, which will be discussed below.

High-precision determinations of fundamental constants allow an additional test of fundamental physics: are such fundamental constants actually constant in time? As there is no physical model explaining the origin of these constants, there is no reason to assume that they do not portray any time-dependent behavior. Such variations could be related to the deep workings of the Universe, as proposed by Dirac [60, 61], but at the very least such variations could point towards new physics. Highprecision spectroscopy is a great tool for testing such effects as there are optical transitions in atoms and molecules available that are very sensitive to variations in the fine structure constant  $\alpha$  or the protonto-electron mass ratio  $m_p/m_e$  [62, 63]. These variations can be tested with high frequency precision in the laboratory with e.g. atomic clocks [64, 65] or compared to astrophysical data which provides a long lookback time to observe a temporal variation in  $m_p/m_e$  [66, 67], or  $\alpha$  [68], or possibly even observing a spatial variation in  $\alpha$  [69–72]. Alternatively, agreement between theory and experiment can also provide constraints on any possible extensions to the Standard Model [73].

#### Fine structure constant

In helium the long-standing testing ground for the fine structure constant is the fine structure splitting of the 2  ${}^{3}P_{0,1,2}$  states as suggested in 1964 [74]. Although the theoretical developments are more complicated in helium than they would be in hydrogen, the lifetimes of the  ${}^{3}P$ states in helium are over two orders of magnitude longer than for the  ${}^{2}P$ states in hydrogen and therefore allow for more accurate experimental determinations.

For almost 50 years spectroscopic measurements have been ongoing to determine the fine structure splitting with currently (sub-)kHz accuracy (e.g. Refs. [75–78] and see Ref. [79] for an overview) using various techniques such as atomic beam optical spectroscopy, microwave spectroscopy with separated oscillatory fields, and saturated absorption spectroscopy. The fine structure splitting is determined with relative uncertainties in the range  $10^{-7} - 10^{-8}$  [79], and combined with theory can give a value of the fine structure constant with  $< 3 \times 10^{-8}$  accuracy [80, 81]. However, the individual experimental results and theory are not accurate enough to provide a single value and are therefore not yet used for determining a competitive value of the fine structure constant [82]. The scattering of the individual experimental results could be related to quantum-interference effects of far off-resonant states, which can slightly change the line shape of the observed transition. Correcting for this effect leads to better agreement between the fine structure results [79], indicating that future experiments which take this effect into account could reach  $< 10^{-8}$  precision in the spectroscopy and as a result  $\sim 10^{-9}$ precision in the fine structure constant determination. At this level the helium experiments become competitive with the electron anomalous magnetic moment experiments [20, 21] to determine the fine structure constant. The above experiments all rely on accurate QED calculations of the energy levels, and therefore an experimental determination of the fine structure constant is never independent from theory.

A QED-independent determination of the fine structure constant is possible using atom interferometry. Already in the original work by Sommerfeld [8] the fine structure constant is written as

$$\alpha^2 = \frac{2R_\infty}{c} \frac{h}{m_e},\tag{1.3}$$

where  $R_{\infty}$  is the Rydberg constant, c the speed of light, h Planck's constant, and  $m_e$  the mass of the electron. This definition did not change even in the relativistic quantum mechanical definition of Dirac [9]. For an atom with mass M, we can rewrite this relationship as

$$\alpha^2 = \frac{2R_\infty}{c} \frac{M}{m_u} \frac{m_u}{m_e} \frac{h}{M},\tag{1.4}$$

where  $M/m_u$  and  $m_e/m_u$  are the atom and electron mass in atomic mass units, respectively. Using atom interferometry the atom recoil velocity  $v_{\rm rec}$  ( $v_{\rm rec} = \hbar k/M$ , which is proportional to h/M) can be measured and allows a QED-independent determination of  $\alpha$ . The Rydberg constant is known with  $5.9 \times 10^{-12}$  precision [82] and  $m_e/m_u$  was recently determined (again using high-precision spectroscopy combined with QED [83]) to  $3 \times 10^{-11}$  relative precision [82]. The accuracy of the factor  $M/m_u$  depends on the atomic species used. Current interferometry experiments have been performed with rubidium or caesium [84–87], of which  $M/m_u$  is known with  $8 \times 10^{-11}$  and  $7 \times 10^{-11}$  relative accuracy, respectively [88].

The most accurate determination to date, using rubidium, determines the ratio h/M with  $1.3 \times 10^{-9}$  precision [85] and therefore  $\alpha$  with  $6.6 \times 10^{-10}$  precision. This provides an important (nearly) QED theoryindependent check with the electron anomalous magnetic moment measurements (and both results are in agreement). Helium is an interesting candidate for these measurements as the atomic mass  $M/m_u$  is known with better precision  $(1.6 \times 10^{-11} [88])$  than rubidium or caesium and therefore provides 'more room at the bottom' [89, 90]. Furthermore, due to the very low second order Zeeman shift in the 2  ${}^{3}S_{1}$  ( $m_{J} = 0$ ) state (see Chapter 5) the magnetic field sensitivity (one of the limiting factors in atom interferometry experiments) is much less.

#### Size of the helium nucleus

Generally calculations ignore the size of the nucleus of an atom as its volume is  $10^{15}$  times smaller than the volumetric extent of the wave-function of the electron. This can be a good approximation but the nucleus, even for the smallest atom, has a finite size and the electron wavefunction will have a finite overlap with the 'inside' of the nucleus. Outside of the nucleus the interaction potential between an electron and the nucleus is governed by Coulomb's law, but inside of the nucleus there can be any sort of charge distribution leading to an energy shift to the eigenenergy of the electronic state which is usually called the 'finite nuclear size effect'.

In the state-of-the-art calculations of level energies for the hydrogen atom (e.g. Ref. [91]) the leading uncertainty in the results is given by the uncertainty in the known size of the hydrogen nucleus (i.e. the proton). If high-precision spectroscopy surpasses the accuracy of the theory, a determination of the size of the proton is possible, and this is exactly what was done up to 2009 [92]. By combining the results of multiple optical transitions that were measured in hydrogen, the Rydberg constant (determining the energy scale of the system) could be determined and the charge radius of the proton could be determined as the remaining variable. Combined with electron-proton scattering data the proton charge radius was determined with 0.78% accuracy [93].

The finite nuclear size effect is proportional to the spatial overlap of the electron wavefunction and the nucleus. As the spatial extent of the wavefunction scales with  $m_e^{-3}$ , substitution of the electron by a muon  $(m_{\mu}/m_e \approx 207 \ [82])$  will increase the finite nuclear size effect by a factor  $8 \times 10^6$  and would be much easier to measure in muonic hydrogen. Once the measurement results were published in 2010 there was a big problem: the proton size determined from muonic hydrogen spectroscopy was  $5\sigma$  smaller than the accepted value [94], and this discrepancy grew to  $7\sigma$  after a second transition was measured in muonic hydrogen [95]. Recently a similar  $(7.5\sigma)$  discrepancy was observed in muonic deuterium measurements [96]. This 'proton radius puzzle' has drawn a lot of attention, as a straightforward explanation of this discrepancy has been ruled out and much effort has turned to further investigation (see e.g. Refs. [97, 98]).

#### 1. INTRODUCTION

For atomic helium direct optical transition frequency metrology cannot be used to determine the absolute size of the helium nucleus as the calculations are not accurate enough to be sensitive to the finite nuclear size. For instance, the finite nuclear size effect on the total ionization energy for the 1  ${}^{1}S_{0}$ , 2  ${}^{1}S_{0}$ , and 2  ${}^{3}S_{1}$  states is 30 MHz, 2.0 MHz, and 2.6 MHz respectively [26], but the total accuracy of the theoretical ionization energies are 36 MHz, 1.9 MHz, and 2.6 MHz [26]. One solution is to perform spectroscopy in He<sup>+</sup> as this is a hydrogen-like system for which theory is again accurate enough, but the wavelengths required for optical spectroscopy are very short making such experiments very challenging. Nonetheless work towards spectroscopy in He<sup>+</sup> is ongoing at MPQ Garching in Germany and at the LaserLaB in Amsterdam. Spectroscopy experiments have been performed in  $\mu^4 \text{He}^+$  and  $\mu^3 \text{He}^+$ (a helium nucleus with a single muon), and the projected accuracy in the nuclear charge radius determination is  $3 \times 10^{-4}$  (0.5 attometer!) [99, 100]. No absolute values are yet presented as the theory to calculate the energy levels of such muonic ions is still under development, and the uncertainties are dominated by nuclear polarizability contributions [101, 102].

Although the absolute ionization energies of the low-lying neutral helium states are known to  $\sim$  MHz precision from theory (see Table 1.1). the *isotope shift* of transitions can be calculated with sub-kHz precision as the mass-independent QED terms largely cancel [103, 104]. This provides a nuclear charge radius difference between two isotopes which can be compared to the aforementioned experiments with muonic helium ions involving the same isotopes. There are currently three measurements of <sup>3</sup>He-<sup>4</sup>He isotope shifts that are accurate enough to be used for nuclear charge radius difference determinations. The first two measurements are done on the 2  ${}^{3}S_{1} - 2 {}^{3}P_{0,1,2}$  transitions by Shiner and coworkers [105] and by Cancio Pastor and coworkers [36, 44]. The third measurement was done on the 2  ${}^{3}S_{1} - 2 {}^{1}S_{0}$  transition in our group by Van Rooij and coworkers [31, 106]. The results on the squared nuclear charge radius difference  $\delta r^2$  are shown in Figure 1.2, which also shows the variation in the nuclear charge radius difference caused by reevaluation of the theory [27, 28, 107].

The current accuracy in the nuclear charge radius difference determined in our group is  $\sigma_{\delta r^2} = 0.011 \text{ fm}^2$ , or  $\sqrt{\sigma_{\delta r^2}} \approx 5 \text{ am} [27, 31]$ . This is



**Figure 1.2** – Squared <sup>3</sup>He-<sup>4</sup>He nuclear charge radius difference  $\delta r^2$  based on the experiments by Van Rooij et al. [31], Cancio Pastor et al. [44], and Shiner et al. [105]. The red results are based on the isotope shift theory in 2012 [44, 107], the blue results based on a reevaluation in 2015 [27]. The black results are based on a reevaluation of the isotope shift on the 2  ${}^{3}S_{1} - 2 {}^{3}P_{0,1,2}$  transition in 2016 [28]. For comparison scattering experiment results are also shown [108].

an order of magnitude larger than the expected accuracy of the results from the  $\mu^4$ He<sup>+</sup> and  $\mu^3$ He<sup>+</sup> results. In order to provide a competitive accuracy in the nuclear charge radius difference the measurement of the 2  ${}^3S_1 - 2 {}^1S_0$  transition frequency has to be significantly improved. Important steps towards an improved measurement of this transition involve narrowing and better understanding of the line shapes of trapped quantum degenerate gases (the subject of Chapter 4) as well as better control or elimination of systematic shifts on the transition frequencies (Chapter 5). A new measurement can also shed more light on the discrepancy between the nuclear charge radius difference results as shown in Figure 1.2.

Nuclear charge radius measurements are also done in heavier (unstable)

isotopes of helium (<sup>6</sup>He and <sup>8</sup>He) to investigate the nuclear few-body physics behind these so-called 'halo' nuclei [109]. As the nuclear charge radii are determined relative to <sup>4</sup>He, a better determination of the <sup>4</sup>He nuclear charge radius would also improve the comparison between theory and experiment for these systems. An extensive recent overview of this work, including the 'proton radius puzzle'-related work in hydrogen and helium, can be found in Ref. [110].

#### 1.5 Physics with ultracold He<sup>\*</sup>

Up until now I have focused on using helium as a testing ground for QED using spectroscopy i.e. as a testing ground for matter and radiation. However, helium (and specifically helium in the metastable  $2 {}^{3}S_{1}$  state, He<sup>\*</sup>) is also a benchmark system for testing atomic physics and quantum mechanics. Its simple electronic structure allows *ab initio* calculations of the molecular potentials of He<sup>\*</sup>-He<sup>\*</sup> collisions, and the internal energy of 19.82 eV of He<sup>\*</sup> atoms allows for single-atom detection on a microchannel plate (MCP) detector or a delay-line detector. This opens the path for quantum optics experiments at a single-atom level that is not easy to reach using the conventional ultracold gases (generally alkali-metal or earth-alkaline atomic species).

Quantum atom optics experiments became feasible with the advent of laser cooling of atoms in the 1980's [111–114]. Once it was possible to optically decelerate and even stop atoms, advances in magneto-optical and magnetic trapping allowed clouds of atoms to be cooled to such low temperatures [115] that new regimes of physics became accessible. Not much later the first Bose-Einstein condensates were observed [116, 117] followed by degenerate Fermi gases [118–120]. This opened a whole new world of physics where many-body interactions can be investigated in highly controlled environments [121] and atomic interactions can even be tuned using Feshbach resonances [122].

Soon also Bose-Einstein condensation of  ${}^{4}\text{He}^{*}$  was achieved at the Institute d'Optique [123] and at the ENS in Paris [124], and later at the VU in Amsterdam [125], ANU in Canberra [126], and MIT in Cambridge [127]. Recent years have seen additional helium BEC experiments at the IQOQI institute at the university of Vienna [128], Institute d'Optique [129], and Amsterdam [130]. For the fermionic isotope <sup>3</sup>He<sup>\*</sup> only in Amsterdam quantum degeneracy was achieved [131]. Currently the groups at the Institute d'Optique, VU, ANU, and Vienna are actively working with ultracold He<sup>\*</sup>. An overview of the experimental techniques and investigations with He<sup>\*</sup> is given in Ref. [132].

#### Polarizabilities

The availability of very accurate *ab initio* transition frequencies and oscillator strengths of helium (see, e.g. Ref. [56]) also allows *ab initio* calculation of the dc and ac polarizabilities of the <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> states without using advanced calculation techniques [133], although such methods can be employed to achieve higher accuracy [134, 135] to put a constraint on the oscillator strengths. We have calculated the ac polarizabilities of <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> for a broad wavelength range (see Chapter 5) in order to build a so-called 'magic wavelength' trap. A recent measurement of a tune-out wavelength (wavelength at which the ac polarizability is zero) of <sup>4</sup>He<sup>\*</sup> [136] is the first experimental test of these calculations.

#### Collisional physics

There is extensive work done on the collisional physics involving He<sup>\*</sup> atoms, both in theory [137–146] and experiment [131, 139, 147–156], as the *ab initio* molecular potentials provide an excellent testing ground. The large internal energy of the He<sup>\*</sup> atoms allows for exothermal loss channels through Penning ionization (He<sup>\*</sup>+He<sup>\*</sup>  $\rightarrow$  He<sup>+</sup>+He+e<sup>-</sup>), which can be studied in these collision experiments. A beautiful example is the determination of the *s*-wave scattering length in the quintet potential of two colliding He<sup>\*</sup> atoms, which was calculated to be 143.0(5)*a*<sub>0</sub> [146] and measured as 142.0(1)*a*<sub>0</sub> [152]. In this thesis (see Chapter 4) the formerly unknown *s*-wave scattering length between a He<sup>\*</sup> atom and 2 <sup>1</sup>S<sub>0</sub> atom is determined and found to be in agreement with *ab initio* calculations [157]. Apart from He<sup>\*</sup>-He<sup>\*</sup> collisions, investigations have expanded to mixtures of He<sup>\*</sup> with alkali-metal atoms, specifically He<sup>\*</sup>-Rb [158–161] as the collisional physics now involves a very large mass ratio between the collision partners.

#### Atom correlations

With the availability of ultracold atoms and single-atom detection, He<sup>\*</sup> is an interesting species for investigation of correlations between atoms. Single-atom detection on a delay-line detector allows full 3D reconstruction of the momentum distribution of a cloud of atoms. Measurements of the second-order correlation functions allowed the first observation of 'bunching' of thermal bosons (also known as the Hanbury Brown-Twiss effect) with massive particles and the disappearance of this effect below the critical temperature of Bose-Einstein condensation [162]. A fruitful collaboration between our group and the group at the Institute d'Optique lead to the first experimental comparison of the Hanbury Brown-Twiss effect for fermions and bosons (displaying antibunching and bunching, respectively) [163]. In the following years fundamental quantum optics theory was confirmed by measurement of higher-order particle correlation functions up to sixth order [164, 165], showing that experiments with massive particles can be very competitive with their purely optical counterparts. Collective effects like superradiance could also be observed by measuring the second-order correlation functions [166, 167], and quantum depletion of a degenerate Bose gas could be observed due to the excellent signal-to-noise ratio with which the momentum distribution of a cloud of He<sup>\*</sup> can be detected [168].

#### Fundamental tests of quantum mechanics

He<sup>\*</sup> is recently used as the test particle for two fundamental experiments displaying quantum effects of massive bosonic particles which were initially performed using (massless) photons. In the first experiment pairs of indistinguishable atoms enter a Mach-Zehnder interferometer [169], and if the atoms are truly indistinguishable they will both simultaneously appear at one of either output ports, but never at both ports simultaneously. This so-called Hong-Ou-Mandel effect is a result of multi-particle interference over all possible paths in the interferomter [170], and this is the first time that this effect is demonstrated for entangled massive bosons.

In the second experiment a single-atom source of He<sup>\*</sup> atoms [171] is used to perform Wheeler's delayed-choice experiment [172]. In this experiment a single atom is sent through a Mach-Zehnder interferometer, but the decision to have an 'open' or 'closed' interferometer (i.e. whether or not the final beamsplitter in the interferometer is employed) is chosen *after* the atom has entered the interferometer. In the 'open' configuration the atom should appear at either port (which is particle-like behavior), and in the 'closed' configuration both ports will display an interference signal (wave-like behavior). It should perhaps appear strange and counterintuitive that a decision at a later time could influence the *a priori* behavior of a particle, but the experiment shows exactly these results in either configuration. This demonstrates that the particle- or wave-like behavior is governed by the quantum description of the whole measurement and not by any (semi-)classical description where the experiment can be described as a series of separate events.

These experiments using He<sup>\*</sup> atoms show how the fundamental principles of quantum mechanics can now be tested with particles that have a finite mass and which move much slower than photons. This is possible because the experimental techniques - single-atom source, single-atom detection - are available for He<sup>\*</sup> atoms but not for many other atomic species.

#### Entangling He<sup>\*</sup> atoms

As He<sup>\*</sup> atoms can be detected with single-atom resolution, they also lend themselves for advanced quantum atom optics experiments involving entangled particles. By colliding two Bose-Einstein condensates of He<sup>\*</sup> a four-wave mixing process takes place that creates atom pairs whose momenta are entangled [173, 174]. The possibility of creating and detecting entangled matter waves allows investigation of fundamental quantum mechanics by observing 'squeezing' and violations of classical behavior [175, 176]. These experiments are clearing the path for advanced tests of quantum mechanics by performing Einstein-Podolsky-Rosen and Belltype experiments [128, 176].

#### **1.6** Outline of this thesis

Helium truly is a workhorse for testing various fields of fundamental physics as it forms a strong combined front with theoretical accuracy and experimental control. In Chapter 2 the experimental setup and techniques used in this thesis are discussed, including a major upgrade of the spectroscopy laser system for the measurement of the  $2 {}^{3}S_{1} - 2 {}^{1}S_{0}$  transition.

As discussed in Section 1.3, QED theory can be directly tested by determining the ionization energy of a state using high-precision spectroscopy. Within this framework, Chapter 3 reports on the first direct measurement of the forbidden  $2 {}^{3}S_{1} - 2 {}^{1}P_{1}$  transition from which the ionization energy of the  $2 {}^{1}P_{1}$  state is determined. This provides a crucial test of QED calculations for this state. Additionally the measurement allows for the most accurate determination of the lifetime of the  $2 {}^{1}P_{1}$  state to date.

The fundamental difference in quantum statistics between fermions and bosons is investigated in Chapter 4, which presents the large differences in line shapes when measuring the 2  ${}^{3}S_{1} - 2 {}^{1}S_{0}$  transition in a degenerate Fermi gas of <sup>3</sup>He<sup>\*</sup> or a Bose-Einstein condensate of <sup>4</sup>He<sup>\*</sup> (which has become possible with the new laser system). This large difference is caused by the fact that fermions (<sup>3</sup>He<sup>\*</sup>) occupy a vastly different spatial and momentum distribution than bosons  $({}^{4}\text{He}^{*})$ , and this measurement therefore is a neat example of how the fundamentally different behavior of bosons and fermions can be observed in optical spectroscopy. The line shapes can be calculated from the theoretical description of the behavior of the quantum degenerate gases, and are shown to be in excellent agreement with the observed line shapes. As the line shape of the Bose-Einstein condensate is partially influenced by the unknown s-wave scattering length of the He<sup>\*</sup> atoms and the excited state  $(2 \, {}^{1}S_{0})$  atoms, a value is determined from the measurements for the first time and is found to be in agreement with new calculations. Surprisingly, the spectrum of the Bose-Einstein condensate reveals a double peak structure, which is explained by the presence of an optical lattice in the optical dipole trap used to confine the atoms.

Finally Chapter 5 contains two major advances for a better measurement of the isotope shift of the 2  ${}^{3}S_{1} - 2 {}^{1}S_{0}$  transition frequency. In the first part so-called 'magic wavelengths' are calculated for which a new optical dipole trap would have a significantly reduced influence on the measured transition frequency. In the second part a new method is proposed for measuring the magnetic field-dependence of the transition frequency shift. The proposed method is based on a two-pulse measurement scheme, known as a Ramsey measurement, and can be insensitive to density-dependent shifts that play a significant role at the current level of accuracy.

# CHAPTER 2

### Experimental setup

The first experiments on laser deceleration, cooling, and trapping of metastable helium atoms in our laboratory started a little over 25 years ago. The current experimental setup originates from that period, and has been under continuous development and improvement ever since. The core vacuum system is the same as used by Rooijakkers [177] to collimate [178], decelerate, and magneto-optically trap metastable helium atoms [179]. Magneto-optically trapped clouds of over  $10^9$  atoms were achieved by Tol and Herschbach [148] at the commonly used He<sup>\*</sup> laser cooling wavelength of 1083 nm, and magneto-optical trapping at the much shorter wavelength of 389 nm [180] or a two-colour (1083 nm + 389 nm) trap [181] was investigated by Koelemeij [182]. The implementation of a magnetic trap [183, 184] and forced evaporative cooling enabled significant progress towards quantum degeneracy of <sup>4</sup>He<sup>\*</sup>. With an improved magnetic trap constructed by Tychkov [185] this culminated in the observation of Bose-Einstein condensation of metastable helium [125].

At the same time progress was made by Stas and McNamara [186, 187] towards cooling and trapping of fermionic <sup>3</sup>He<sup>\*</sup> [188]. Due to the excellent collisional properties between the two isotopes, sympathetic cooling of <sup>3</sup>He<sup>\*</sup> was possible using <sup>4</sup>He<sup>\*</sup> and a mixture of a degenerate Fermi gas and a Bose-Einstein condensate of He<sup>\*</sup> was achieved [131]. The experimental setup had advanced to a point where atomic physics and quantum physics could be investigated with some of the lightest atoms that can be cooled to quantum degeneracy. It allowed for investigation of collisional properties between the isotopes [153, 154] in one of the few systems where the *ab initio* molecular potentials are available.

But also fundamental quantum statistics could be investigated in this experiment, and another major achievement was the fruitful collaboration between the VU and colleagues from the Institute d'Optique in Orsay. A delay-line detector from France was installed in the setup in Amsterdam and used to measure the second order correlation function of an expanding cloud of ultracold <sup>4</sup>He<sup>\*</sup> or <sup>3</sup>He<sup>\*</sup>. As the atoms are either bosons or fermions, they display (anti)bunching due to the (anti)symmetrization requirements on their wavefunctions. This can be observed as an increase or decrease in the second order correlation function of the atoms which can be measured using the delay-line detector. After a short but intense measurement campaign this so-called Hanbury Brown-Twiss effect was for the first time shown in both massive bosons and fermions [163, 189].

The experiment got its most recent major upgrade with the implementation of an optical dipole trap by Van Rooij [190]. The optical dipole trap allowed new degrees of freedom to be addressed, as now not only the  $m_J > 0$  ( $m_F > 0$  for <sup>3</sup>He<sup>\*</sup>) magnetic substates could be trapped but all magnetic substates of <sup>4</sup>He<sup>\*</sup> and <sup>3</sup>He<sup>\*</sup> became available. This allowed investigation of spin-dependent collisions and a search for possible Feshbach resonances [156]. As the optical dipole trap became operational, a new line of research was started by performing high-precision spectroscopy in He<sup>\*</sup>. The optical dipole trap allows for a high degree of control over the atoms and accurate determination of the various systematic effects, which resulted in the measurement of the 2  ${}^{3}S - 2 {}^{1}S$ isotope shift between <sup>4</sup>He<sup>\*</sup> and <sup>3</sup>He<sup>\*</sup> from which the nuclear charge radius difference was determined with 1% accuracy [31].

Indeed the experimental setup has a distinguished service record, and many changes have occurred in its many years of existence. The current status of the experimental setup will be discussed in this chapter, and any relevant changes with respect to the previous thesis [190] will be further elaborated on.

#### 2.1 Slowing, trapping, and cooling He<sup>\*</sup>

#### $\mathrm{He}^*$ source

The design of the source has not changed since the origin of the project in the early '90s [177, 178]. A quartz tube with a tantalum needle inside is fixed to a boron-nitride nozzle, which is mounted in a liquid nitrogen-cooled copper ring. Behind this ring is a skimmer to collimate the beam and allow for differential pumping. Helium gas flows into the quartz tube from the back, and a dc voltage of -2.7 kV is maintained over the tantalum needle and the skimmer to create a plasma discharge. The plasma beam exits through the boron-nitride nozzle, where the velocity of the beam is reduced by a factor of two through collisions with the wall as it is kept at liquid nitrogen temperature. The discharge is typically operated with helium flow and pressure such that the source chamber pressure is  $2 \times 10^{-5}$  mbar and a discharge current of 7 mA is sustained. Under these conditions the liquid nitrogen cooled beam has a most probable velocity of 1100 m/s [177].

After long operation (several months), the quartz tube and tantalum needle are coated with ablated material from the source itself (e.g. the boron-nitride disc). If this is prolonged, the discharge will suffer from sudden interruptions and yearly maintenance is therefore required. This involves cleaning of the quartz tube and removal of deposited materials on the tantalum needle and can be done within an hour of breaking the vacuum, although it will take several hours of running the discharge before it returns to nominal operation.

Recently the liquid nitrogen reservoir was refitted with better isolation material and the typical refill time of the reservoir is now 1,5 hours instead of 45-60 minutes.

#### Collimating and slowing He<sup>\*</sup>

The basis of the experimental setup is shown Figure 2.1. In order to improve the flux of He<sup>\*</sup> in the main chamber the expanding plasma beam is collimated in two orthogonal directions using a curved-wavefront technique [177]. This involves lightly focused collimation beams of which the wavefront curvature is equal to the curvature of the trajectory of



**Figure 2.1** – A schematic overview of the vacuum system. A dc discharge in the source chamber creates a beam of excited helium atoms, which are cooled by a liquid-nitrogen temperature nozzle. The cooled beam is collimated and deflected in a small angle in the collimation section, where a knife-edge (not shown) blocks the non-He<sup>\*</sup> state atoms. When operating with <sup>3</sup>He<sup>\*</sup>, the background gas from the source and collimation chambers is returned into the recycling system. Through the Zeeman slower, the atoms enter the magneto-optical trap (MOT) in the trap chamber. In the current system three large turbopumps (T) are used, three small turbopumps (ST), a helium-proof scroll pump (Sc) and several membrane pumps (M) to provide a low backing pressure. Figure adapted from [190].

the cooled atoms. In this way the atoms remain resonant with the total wavefront, such that continuous radiation pressure can be exerted on the atom beam. A special feature in our setup is a slight imbalance along the horizontal axis of collimation, which causes the beam of He<sup>\*</sup> to be deflected in a small angle. As only He<sup>\*</sup>-state atoms are resonant with the collimation light, this small deflection allows all other atoms to be blocked by a knife-edge. In this way the background load on the main chamber is minimized, which is advantageous for achieving a low background pressure in the main chamber of  $5 \times 10^{-11}$  mbar.

When operating with the rare  ${}^{3}\text{He}^{*}$  isotope, a recycling system containing liquid-nitrogen cooled sodium zeolite molecular sieves is used to recover  ${}^{3}\text{He}^{*}$  atoms which do not pass the knife-edge into the Zeeman slower. The recycling system was only connected to the source chamber, but this has been changed to include both the source and collimation chamber as shown in Figure 2.1. In this way the usage of  ${}^{3}\text{He}^{*}$  has been extended, without compromising the pump capacity of the source and collimation chambers.

After the collimation section, the He<sup>\*</sup> atoms enter the two meter long Zeeman slower facing a counterpropagating circularly polarized laser beam with detuning -250 MHz ( $\sim -154\Gamma$ ) from the 2  ${}^{3}S_{1} - 2 {}^{3}P_{2}$ cycling transition (see Section 2.3) such that the atoms always return to the same magnetic substate. In the Zeeman slower the He<sup>\*</sup> atoms continuously absorb photons with a recoil momentum opposite to their own forward momentum, and the atoms will slow down. As the atoms slow down near the end of the Zeeman slower, the change in Doppler shift  $\Delta \omega_{D} = -\vec{k} \cdot \vec{v}$  is compensated by a spatially changing magnetic field  $\vec{B}$  in the Zeeman slower  $\Delta \omega_{ZS} = \mu_{B} |\vec{B}|$ , where  $|\vec{k}| = 2\pi/\lambda$  is the wave number of the laser beam and  $\mu_{B}$  the Bohr magneton. This ensures that the atoms remain resonant with the laser beam during the deceleration to a final velocity of around 50 m/s, which is sufficiently slow to be captured by the magneto-optical trap.

It is the Zeeman slower section which requires the discharge source to be liquid-nitrogen cooled as the length of the Zeeman slower scales quadratically with the initial velocity of the atomic beam. Hence a factor of two reduction in the initial velocity reduces the required Zeeman slower length to 'only' 2 m. Second, if the polarization of the laser beam is not perfectly circular the magnetic field in the Zeeman slower cannot exceed 563 G, as there is a level crossing between magnetic sublevels which would allow atoms to be pumped to a different state and therefore be lost from the Zeeman slower. Although this would not occur for a perfectly circularly polarized laser beam, this limits the initial velocity of the atomic beam to approximately 1130 m/s, just above the liquid nitrogen-cooled atomic beam velocity of 1100 m/s.

#### Magneto-optical trapping of He<sup>\*</sup>

The decelerated atomic beam enters the main UHV chamber of the setup, where the atoms are trapped in a magneto-optical trap (MOT). Magneto-optical trapping is a well-established technique [113, 114], and combines velocity-dependent laser cooling with spatially-dependent trapping in a magnetic quadrupole potential. The spatial dependence is cre-

ated by the quadrupole magnetic field; as the atoms move away from the center they experience an increasing magnetic field (and Zeeman shift) shifting them in resonance with a circularly polarized counterpropagating laser beam. As the photon absorption increases, the atoms are 'pushed' towards the center of the trap where the magnetic field is smallest ( $|\vec{B}| = 0$  in the center of a simple quadrupole trap). In the center of the trap, where the magnetic field is small, the atoms are cooled by optical molasses.

This is a general description of most MOTs. There are additional complications when working with metastable atoms, as they have a large internal energy that can be released in a collision. This process is explained in the next section, and it limits the densities at which one can operate the MOT as loss rates will become too large. Therefore the MOT operates with three sets of retroreflected beams with a large tophat intensity profile beam diameter of 3 cm, a typical power of 3.5 mW per beam, and a detuning of -35 MHz ( $\sim -22\Gamma$ ) from the  $2^{3}S_{1} - 2^{3}P_{2}$ transition. This is a relatively large detuning for MOTs, but it is necessary to reduce the photo-assisted collision rate between 2  ${}^{3}S_{1}$  and  $2^{-3}P_2$  atoms, of which the two-body loss rate constant is on the order of  $10^{-7}$  cm<sup>-3</sup>/s [147, 151]. The applied magnetic field gradient is  $\partial B/\partial z = 2 \partial B/\partial \rho = 22$  G/cm (corresponding to a MOT coil current of 32 A [185]). For these experimental settings we typically trap  $3 \times 10^8$ atoms at a temperature of 0.56 mK and a peak density of  $\sim 10^9$  cm<sup>-3</sup> with 4 seconds loading time of the MOT. Larger MOTs (with  $> 10^9$ atoms at  $T \approx 1$  mK) were obtained in the past [148], but with the lower temperature in the current configuration we find a better starting condition for BEC production.

#### Penning ionization

This section is a small deviation from the description of the experimental setup, as it is important to discuss the collisional properties of metastable atoms as they have a direct consequence for the design of the setup and the specific cooling and trapping processes.

At low temperatures collisions between two (or more) atoms are no longer described by a hard-sphere collision model and a quantum mechanical approach is required. In the quantum mechanical picture the atoms approach each other in a Born-Oppenheimer molecular potential with quantized angular momentum states l = 0, 1, 2, ... and the full solution of this collision is described as a wavefunction expanded in a series of partial waves [186, 191]. The lowest energy collision, at l = 0 angular momentum, is called an *s*-wave collision (l = 1 is *p*-wave, l = 2 is *d*-wave, etc.). The contribution of the different partial waves decreases as the temperature decreases, and for temperatures below 6.5 mK (10 mK for <sup>3</sup>He<sup>\*</sup> due to its lower mass) [153] only the *s*-wave collisions play a role. In this so-called *s*-wave regime the collisions are characterized by a single parameter: the *s*-wave scattering length *a*. At zero temperature the collision cross-section  $\sigma$  reduces to  $\sigma = 4\pi a^2$  for identical bosons, which is (perhaps surprisingly) very similar to the parametrization of the classical hard-sphere collision model. The main difference is that the scattering length is intimately linked to the exact Born-Oppenheimer potential of the collision instead of the classical 'size' of the atoms.

For an s-wave collision of two He<sup>\*</sup> atoms, the total electron spin per atom ( $\vec{s} = 1$  as the spins are parallel for the 2  ${}^{3}S_{1}$  state) can be added to a total spin of the two atoms  $\vec{S} = \vec{s_{1}} + \vec{s_{2}} = 0, 1, 2$ . These three total spins are associated with the singlet  ${}^{1}\Sigma_{g}^{+}$ , triplet  ${}^{3}\Sigma_{u}^{+}$ , and quintet  ${}^{5}\Sigma_{g}^{+}$  Born-Oppenheimer potentials respectively, depending on the total spin projection  $m_{S}$ . For instance, in a spin-polarized sample of  ${}^{4}\text{He}^{*}$ atoms in the  $m_{J} = +1$  state we have  $m_{S} = 2$  and the collision is solely described by the quintet potential. For a collision between a  $m_{J} = +1$  and  $m_{J} = -1$  atom we have  $m_{S} = 0$  and the collisions happen in the singlet, triplet, and quintet potential. Note that the triplet potential is associated with odd-parity (i.e. *p*-wave) collisions and therefore negligible in the *s*-wave scattering regime.

The electrostatic interactions between the He<sup>\*</sup> atoms allow a coupling of the singlet and triplet potential to other states, thereby releasing a lot of their internal energy (of 19.82 eV each). The two processes through which this can happen are called Penning ionization (PI) and associative ionization (AI):

$$\operatorname{He}^{*} + \operatorname{He}^{*} \to \begin{cases} \operatorname{He} + \operatorname{He}^{+} + \operatorname{e}^{-} & (\operatorname{PI}) \\ \operatorname{He}_{2}^{+} + \operatorname{e}^{-} & (\operatorname{AI}) \end{cases}$$

PI and AI (commonly both are referred to as 'Penning ionization') have both been studied extensively for He<sup>\*</sup> in experiment [139, 148, 153, 155, 156] and theory [137, 138, 141, 143], and they provide an interesting path of understanding ultracold collisions involving exothermal processes. Unfortunately both PI and AI lead to atom loss in a trapped cloud, as the final states can no longer be laser-cooled or trapped. The associated loss rate constant was determined to be  $\sim 1 \times 10^{-10}$  cm<sup>3</sup>/s for an unpolarized gas of He<sup>\*</sup> [148, 155]. Penning ionization therefore severely limits the possibilities of cold atom experiments, and it is for this reason that it was long considered impossible to cool metastable atoms to even lower temperatures to achieve quantum degeneracy.

Collisions for spin-polarized atoms in the  $m_J = \pm 1$  states occur in the pure quintet potential, which is a special case as this potential does not couple to the PI and AI decay channels which would require a (forbidden) spin-flip. Of course there are higher-order processes that occur such as relaxation-induced Penning ionization and magnetic field-dependent spin-relaxation [141, 156], but typically the two-body loss rate in the quintet potential is ~  $10^{-14}$  cm<sup>3</sup>/s, four orders of magnitude less than PI and AI.

Apart from two-body collisions of He<sup>\*</sup> atoms, photo-assisted collisions also play a role in a MOT. The detuned light in the MOT couples the 2  ${}^{3}S_{1}$  and 2  ${}^{3}P_{2}$  atoms to the molecular potentials for two He<sup>\*</sup> atoms, of which the singlet and triplet potentials then allow Penning ionization and the atoms are lost from the MOT [150]. The two-body loss rate constant is on the order of  $10^{-7}$  cm<sup>3</sup>/s [147, 151], three orders of magnitude larger than the Penning ionization loss channel. At typical peak densities of  $10^{9}$  cm<sup>-3</sup> the lifetime of the cloud is  $\ll 1$  s and much shorter than the one-body lifetime due to the background pressure of the main chamber of tens of seconds. Fortunately this loss rate can be suppressed by choosing a large detuning from the  $2 {}^{3}S_{1} - 2 {}^{3}P_{2}$  transition, at a cost of a higher MOT temperature. By choosing relatively large MOT beam diameters, the large volume of the cloud still allows trapping of an appreciable number of atoms at a lower density.

#### Magnetic trapping and evaporative cooling

Neutral atoms can have a magnetic moment which in the presence of a magnetic field *B* leads to a potential energy  $U = \mu_B g_J m_J B$  for <sup>4</sup>He<sup>\*</sup> and  $U = \mu_B g_F m_F B$  for <sup>3</sup>He<sup>\*</sup>, where  $\mu_B = 1.4$  MHz/Gauss is the Bohr

magneton and  $g_J \approx 2.002$  and  $g_F \approx 1.335$  the Landé factors for the metastable states (F = 3/2 for <sup>3</sup>He<sup>\*</sup>) [186]. An appropriate magnetic field geometry can thus be used to trap atoms (in the  $m_J = +1$  or  $m_F = +3/2, +1/2$  states) without using light.

After loading the MOT, the atoms are transferred to the magnetic trap (MT) so they can be cooled to quantum degeneracy. A spin-polarizing beam is applied for 2 ms when switching from the MOT to the MT configuration to pump the atoms to the magnetically trappable  $m_J = +1$  state ( $m_F = +3/2$  for <sup>3</sup>He<sup>\*</sup>). This has the advantage of increasing the atom number in the MT (as the atoms are distributed over all magnetic substates in the MOT), and the spin-polarized gas is much more stable against Penning ionization as discussed in the previous section.

The MT cannot simply be a quadrupole magnetic field which has a zero crossing of the magnetic field in its center. If the spin-polarized atoms move through the center, the zero magnetic field would lead to depolarization (as there is no well-defined quantization axis any more), and the atoms would immediately be lost from the trap or even Penning ionize. This loss of atoms - called Majorana loss [192–194] - is more likely to happen to atoms that spend more time near the bottom of the trap. As a result the colder atoms from the total distribution are lost faster than the hotter ones effectively leading to heating of the cloud. Furthermore, the Majorana heating rate scales inversely proportional with mass, which does not help when working with helium. This is a serious limitation for cooling atoms to lower temperatures, although solutions have been achieved with quadrupole magnetic traps in combination with optical dipole beams [130].

The Majorana loss issue is solved by using a Ioffe-Pritchard cloverleaf trap, of which our currently used trap was designed and installed by Tychkov [125]. This configuration provides a trapping potential with a non-zero magnetic field in the center so atoms always experience a quantization axis and Majorana losses are prevented. The cloverleaf trap exists of six pairs of coils: two concentric pairs of pinch and compensation coils, and four pairs of radial confinement coils (see [185] for details on the design). The pinch and compensation coils are operated in Helmholtz configuration in the MT, and the compensation coils are used in anti-Helmholtz configuration for the MOT to create the required quadrupole magnetic field. Once the atoms are transferred to the MT (with a loss of 25-40% in the atom number, depending on the exact loading conditions) the temperature has increased to  $\sim 1 \text{ mK}$  due to the optical pumping and the increased density. Therefore a single, weak, red detuned beam is used to Doppler cool the atoms for 2 seconds along the long axis of the trap. Although this is done along a single axis (i.e. in 1D), the reabsorption of red detuned photons within the cloud will lead to cooling in all directions of the cloud [125, 195]. Without much variation a temperature of  $\sim 0.13$  mK is typically reached (which is approximately three times the Doppler limit temperature  $T_{\text{Dop}}$ , see Table 2.1 in Section 2.3). As this process depends on the optical thickness of the cloud, it generally shows a threshold-like behavior; it works properly over a broad range of atom numbers, but as soon as the atom number is too low it simply will not work any more. As a rule of thumb over  $2 \times 10^8$  atoms are needed in the MOT in order to have sufficient atoms for this so-called 1D Doppler cooling.

After the 1D Doppler cooling step the MT is compressed by reducing the current through the compensation coils. This lowers the magnetic field in the trap center and increases the magnetic field curvature, thereby increasing the density and collision rate of the atoms. At this stage the atoms can be cooled to degeneracy by forced rf-induced evaporative cooling. The rf induces cascaded transitions from the  $m_J = +1$  states to the untrappable  $m_J = 0, -1$  states, and by slowly ramping from a large rf frequency (50 MHz) to a low frequency (4.0 MHz) the hotter atoms are expelled from the trap and the cloud rethermalizes to a lower temperature. Using a ramp of 6 seconds we obtain a quantum degenerate gas in the magnetic trap. After transfer to the optical dipole trap (see next section) we are left with ~ 10<sup>6</sup> atoms predominantly in a Bose-Einstein condensate and a temperature of ~ 0.2  $\mu$ K of the thermal fraction.

When working with <sup>3</sup>He<sup>\*</sup> this procedure cannot be used as the atoms stop thermalizing once the temperature is below the *p*-wave barrier of 10 mK [153]. Fortunately there are *s*-wave collisions possible between <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> atoms with an *s*-wave scattering length of 27.18(45)  $a_0$ [131, 146], which allows for efficient sympathetic cooling. In practice this means that a MOT of <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> is loaded and the whole experimental procedure is performed as explained for <sup>4</sup>He<sup>\*</sup>, resulting
in a thermalized quantum degenerate  ${}^{3}\text{He}^{*}$ - ${}^{4}\text{He}^{*}$  mixture. A pure  ${}^{3}\text{He}^{*}$  cloud in the MT can be achieved by using the rf-ramp to cut away the  ${}^{4}\text{He}^{*}$  atoms<sup>1</sup>. In the dipole trap resonant imaging light is used to blast the  ${}^{4}\text{He}^{*}$  atoms away, and depending on the loading conditions over  $10^{6}$   ${}^{3}\text{He}^{*}$  atoms can remain in the dipole trap at  $T \approx 0.2 \ \mu\text{K}$ . In the experiments described in Chapter 4 the  ${}^{3}\text{He}^{*}$  loading process was not fully optimized and we typically loaded  $\sim 3 \times 10^{5}$  atoms at  $T \approx 0.2 \ \mu\text{K}$ .

## Optical dipole trap

Magnetic trapping of neutral atoms was a giant leap forward in achieving quantum degenerate gases, but it did not allow for trapping of field-insensitive or high-field seeking states (i.e. the  $m_J = 0, -1$  and  $m_F = -1/2, -3/2$  states). Furthermore, magnetic field gradients can lead to unwanted systematic shifts in experiments during the measurement itself or by transient fields when switching off the MT. This changed with the advent of the optical dipole trap.

As optical dipole traps work with laser beams, they allow great spatial and temporal flexibility [196] because the trapping no longer depends on magnetic substates but on the wavelength and intensity of the laser beams. Apart from various geometries, interference between different laser beams can be used to engineer a periodic potential to investigate ultracold atom physics in optical lattices [121, 197].

### Basic physics of optical dipole traps

The basic operation of an optical dipole trap is based on the induced oscillating electric dipole  $\vec{d}(\omega)$  of an atom placed in an oscillating electric field  $\vec{E}(\omega)$ . The induced dipole and the electric field are related as  $\vec{d}(\omega) = \alpha(\omega)\vec{E}(\omega)$ , where  $\alpha(\omega)$  is called the polarizability. The polarizability generally is a complex number, as it depends on the oscillation frequency  $\omega$  of the field and on the relative phase of the oscillating dipole with respect to the field. For a classical damped driven harmonic oscillator model with driving frequency  $\omega$ , resonance frequency  $\omega_0$  and

 $<sup>^1{\</sup>rm This}$  is possible because the Zeeman splitting for the rf transitions is 2.8 MHz/G for  $^4{\rm He}^*$  and 1.9 MHz/G for  $^3{\rm He}^*.$ 

damping rate  $\Gamma$  (also known as the linewidth), the polarizability is [196]

$$\alpha(\omega) = 6\pi\epsilon_0 c^3 \frac{\Gamma/\omega_0^2}{\omega_0^2 - \omega^2 - i\Gamma(\omega^3/\omega_0^2)}.$$
(2.1)

The time-averaged interaction energy is

$$U_{\rm dip}(\vec{r}) = -\frac{1}{2} \langle \vec{d} \cdot \vec{E} \rangle = -\frac{1}{2\epsilon_0 c} \operatorname{Re}(\alpha(\omega)) I(\vec{r}), \qquad (2.2)$$

with the intensity of the field defined as  $I(\vec{r}) = (\epsilon_0 c/2) \times \langle |\vec{E}(\omega, \vec{r})|^2 \rangle$ . The dipole potential and the off-resonant scattering rate for a two-level system scale as [196]

$$U_{\rm dip} \propto \left(\frac{\Gamma}{\Delta}\right) \frac{I(\vec{r})}{\omega_0^3},$$
 (2.3)

$$\Gamma_{\rm scatt} \propto \left(\frac{\Gamma}{\Delta}\right)^2 \frac{I(\vec{r})}{\omega_0^3},$$
(2.4)

where  $\Gamma/2\pi$  is the linewidth of the transition, and  $\Delta = \omega - \omega_0$  the detuning from the resonance frequency. The polarizability needs to be positive in order to have a trapping potential  $(U_{\rm dip} < 0 \text{ for } \alpha > 0)$ at an intensity maximum, which holds for red detunings  $(\omega < \omega_0)$ . Blue detuned light  $(\omega > \omega_0)$  will create an antitrapping potential at an intensity maximum, which can be used in more complex configurations [196]. Furthermore, the scattering rate decreases faster with increasing  $\Delta$  than the dipole potential. This is the reason why trapping in far off-resonant light fields is possible at all.

Although the basic principles are the same, an accurate description of the polarizability of an atom requires a bit more detail as one needs to take the polarization of the light into account and therefore the relative transition strengths involved. This is especially needed if there are multiple optical transitions contributing to the polarizability. This is explained in Chapter 5 for extended calculations [133] and just briefly discussed here for the purpose of trapping He<sup>\*</sup> atoms. Using the LS coupling scheme, the expression for the polarizability of a 2  ${}^{3}S_{1}$  ( $m_{J} = +1$ ) state atom in a light field with polarization state q and angular frequency  $\omega$  is

$$\alpha(q,\omega) = 6\pi\epsilon_0 c^3 \sum_{n'} \sum_{J'} (2J'+1) \begin{pmatrix} 1 & 1 & J' \\ -1 & q & m'_J \end{pmatrix}^2 \times \frac{A_{n'J'}}{\omega_{n'J'}^2(\omega_{n'J'}^2 - \omega^2)},$$
(2.5)

where we sum over all transitions 2  ${}^{3}S_{1}$   $(m_{J} = +1) \rightarrow n' {}^{3}P_{0,1,2}$  for each fine structure manifold, and the resonance frequency is  $\omega_{n'J'}$  with Einstein A coefficient  $A_{n'J'}$ . The polarization state q = -1, 0, +1 of the electric field determines if we have  $\sigma^{-}, \pi$ , or  $\sigma^{+}$  transitions contributing, and the relative weights of the transitions are given by the 3j symbol between the big brackets.

The great advantage of working with helium is that *ab initio* transition frequencies and Einstein coefficients are readily available for transitions up to n = 10 [56]. We have used this data to calculate the polarizability of <sup>4</sup>He<sup>\*</sup> and <sup>3</sup>He<sup>\*</sup> for a broad wavelength range in our search for magic wavelength candidates (see Chapter 5). At the operating wavelength of 1557.3 nm of our optical dipole trap and for linear polarized light (q = 0) we find that the polarizability of <sup>4</sup>He<sup>\*</sup> is

$$\alpha(\lambda = 1557.3 \,\mathrm{nm}) = 604 \,a_0^3 = 9.96 \times 10^{-39} \,\mathrm{JV}^{-2}\mathrm{m}^2.$$
 (2.6)

In this far off-resonant regime (the nearest transition is  $2 {}^{3}S_{1} \rightarrow 2 {}^{3}P_{J}$  at 1083 nm) the polarizability is the same for  ${}^{3}\text{He}^{*}$ , which means that the isotopes experience the same optical dipole potential.

#### A single-beam optical dipole trap

The simplest case for optical trapping is a single focused laser beam. The intensity profile of a  $\text{TEM}_{00}$  mode laser beam with total power P propagating along the z-axis is

$$I(r,z) = \frac{2P}{\pi w^2(z)} \exp\left(-\frac{2r^2}{w^2(z)}\right),$$
(2.7)

where r is the radial coordinate and the  $1/e^2$  intensity profile radius  $w(z) = w_0 \sqrt{1 + (z/z_R)^2}$ , where the minimum waist  $w_0$  is called the beam waist and  $z_R = \pi w_0^2 / \lambda$  the Rayleigh length. The peak intensity

is defined as  $I_0 = 2P/\pi w_0^2$ . The dipole potential corresponding to this intensity field can be calculated using Eq. 2.2, resulting in trap depth  $U_0 = \alpha I_0/2\epsilon_0 c$ . To understand the basic properties of this trap, the potential around the center of the trapping potential is expanded using the harmonic approximation:

$$U_{\rm dip} \approx -U_0 \Big[ 1 - 2 \Big( \frac{r}{w_0} \Big)^2 - \Big( \frac{z}{z_R} \Big)^2 \Big].$$
 (2.8)

The corresponding axial (longitudinal) and radial (transverse) trapping frequencies are

$$\omega_z = \sqrt{\frac{2U_0}{mz_R^2}},\tag{2.9}$$

$$\omega_r = \sqrt{\frac{4U_0}{mw_0^2}},\tag{2.10}$$

and the aspect ratio of the trap is  $\omega_r/\omega_z = \sqrt{2\pi}w_0/\lambda \approx 240$  for a beam waist of  $w_0 = 85 \ \mu \text{m}$  and wavelength  $\lambda = 1557.3$  nm as used in our experiment. This large aspect ratio is no surprise as the Rayleigh length  $z_R \approx 1.5$  cm is large compared to the beam waist. A more urgent problem is the fact that this Rayleigh length is comparable to the distance (3.3 cm) between the windows of the vacuum chamber, and trapped atoms will simply hit the windows if we trap them in a single beam optical dipole trap. For magnetically trappable atoms this problem could be solved by applying a magnetic quadrupole field to provide more confinement. This is shown to work for He<sup>\*</sup> [130, 198], but it provides serious systematic shifts when doing high-precision spectroscopy. Therefore a crossed-beam geometry was chosen to trap the atoms.

#### A crossed-beam optical dipole trap

The large aspect ratio of a single-beam optical dipole trap is reduced by overlapping a refocused laser beam at an angle  $\theta = 19^{\circ}$  with the original beam. A schematic view of this trap is shown in Figure 2.2. The two laser beams have orthogonal linear polarizations in order to minimize interference of the light fields which could create an optical



Figure 2.2 – Schematic view of the crossed-beam optical dipole trap with respect to the orientation of the MOT coils (part of the Ioffe-Pritchard trap). A f = 400 mm lens ('L') focuses the incoming beam in the vacuum chamber, after which it is collimated and the polarization rotated by 90° using a half-wave plate ('HWP') and a polarizing beam splitter ('PBS') to prevent any interference effects that could lead to an optical lattice. The beam is refocused at an angle  $\theta = 19^{\circ}$  with respect to the first beam after which it is collected by a beam dump or a power meter. The MCP detector is placed 17 cm below the center of the trap.

lattice. Defining the long axis of the potential as the z-axis and the out-of-plane vertical axis as the x-axis, the trapping potential can now be approximated as

$$U_{\rm dip} \approx -U_0' \Big[ 1 - 2 \Big( \frac{z}{w_z} \Big)^2 - 2 \Big( \frac{y}{w_y} \Big)^2 - 2 \Big( \frac{x}{w_0} \Big)^2 \Big], \tag{2.11}$$

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where the trap depth  $U'_0 = 2U_0$  because we have two beams<sup>2</sup>. The trap waists are now defined as [196]

$$w_z^2 = \left[ \left( \frac{\sin(\theta/2)}{w_0} \right)^2 + \left( \frac{\cos(\theta/2)}{2z_R} \right)^2 \right]^{-1} \approx \left( \frac{w_0}{\sin(\theta/2)} \right)^2, \qquad (2.12)$$

$$w_y^2 = \left[ \left( \frac{\cos(\theta/2)}{w_0} \right)^2 + \left( \frac{\sin(\theta/2)}{2z_R} \right)^2 \right]^{-1} \approx \left( \frac{w_0}{\cos(\theta/2)} \right)^2.$$
(2.13)

The final approximation is valid to four digits due to the large difference between  $z_R$  and  $w_0$ . Using these approximations the harmonic trap frequencies are

$$\omega_x = \sqrt{\frac{4U_0'}{mw_0^2}},$$
 (2.14)

$$\omega_y = \omega_x \, \cos(\theta/2), \qquad (2.15)$$

$$\omega_z = \omega_x \, \sin(\theta/2). \tag{2.16}$$

The crossed-beam dipole potential still has radial symmetry as  $\omega_y/\omega_x \approx 0.95$ , but the axial-radial aspect ratio has now decreased from the single-beam aspect ratio of 240 to  $\omega_y/\omega_z \approx 6.0$ . Hence the crossed-beam dipole trap provides much more spatial confinement than a single-beam trap.

### Loading atoms in the crossed-beam optical dipole trap

The center of the optical dipole trap should ideally be aligned to exactly overlap with the center of the magnetic trap, such that the evaporatively cooled atoms transfer efficiently into the optical dipole trap. The alignment of the optical dipole trap is optimized such that the transfer efficiency of the number of atoms is optimal, but as a result the optical dipole trap is slightly misaligned with respect to the magnetic trap center. This is caused by transient magnetic field gradients which 'push' the atoms out of the center of the trap when switching off the magnetic trap. This is also visible as a distortion in the time-of-flight signals, but after a hold time of 500 ms in the optical dipole trap the cloud is

<sup>&</sup>lt;sup>2</sup>In practice  $U'_0 < 2U_0$  due to various reflection losses.

thermalized properly again. When working with  ${}^{4}\text{He}^{*}$  this is our starting point for measurements, but when working with  ${}^{3}\text{He}^{*}$  a  ${}^{3}\text{He}^{*}$ - ${}^{4}\text{He}^{*}$  mixture is loaded in the optical dipole trap to make sure that the  ${}^{3}\text{He}^{*}$  atoms are thermalized again. After thermalization the  ${}^{4}\text{He}^{*}$  atoms are blasted away with imaging light which is far off-resonant for the  ${}^{3}\text{He}^{*}$  atoms.

The optical dipole trap has not changed much since the previous thesis [190], although some changes in its use have been implemented. In the original spectroscopy scheme [31] the 2 W commercial fiber laser system (NP Photonics Scorpio) had a dual function of supplying light for the optical dipole trap and light for performing spectroscopy on the  $2^{3}S - 2^{1}S$  transition and therefore the fiber laser frequency was locked to the frequency comb. Since then we have acquired and implemented a new spectroscopy laser system (see Section 2.5) and only use the free running NP Photonics fiber laser system, properly detuned from resonance, for the optical dipole trap. The He\*Rb experiment - neighbors in our laboratory - pick off a few mW of the laser light to seed their own amplifier system for an optical dipole trap [130, 199].

To make the dipole trap power more versatile, a  $\lambda/2$  waveplate was mounted in a motorized rotation stage in combination with a polarizing beam splitter to allow digital control over the optical dipole trap power. As we recently noticed drifts in the optical dipole trap power of  $\sim 15\%$ over the course of an hour, a slow digital feedback loop to the rotation stage was implemented by digital acquisition of a photodiode signal and digital computation of the correction to the rotation stage angle after every measurement. As the transmission of the polarizing beam splitter can be measured as function of the rotation stage angle, one can calculate the local derivative of the power with respect to the angle  $\partial P/\partial \theta$ and use it to estimate the angular correction  $\Delta \theta = \Delta P / (\partial P / \partial \theta)$  for a measured differential power  $\Delta P$  with respect to the setpoint. This linearization assumes that the angular corrections are small, as is the slow drift that is observed. Empirical optimization of the PI settings and the time constant for integration provides a long term stability of  $\sim 0.3\%$ of the signal<sup>3</sup>. With the drift in the optical dipole trap - and thereby

<sup>&</sup>lt;sup>3</sup>Long term stability is defined here as the standard deviation  $\sigma$  of the dataset divided by the average  $\mu$  i.e.  $\sigma/\mu$ , which is also known as the coefficient of variation.

the varying ac Stark shift on the transition frequency - eliminated, interpretation of the measured line shapes is much more straightforward. In most experiments ~ 240 mW is sent into the UHV chamber, which adds to a combined power of both trapping beams<sup>4</sup> of ~ 390 mW. With a beam waist of  $w_0 = 85 \ \mu\text{m}$  a trap depth of  $U'_0/k_B \approx 4.5 \ \mu\text{K}$  is estimated with corresponding trap frequencies ( $\omega_x, \omega_y, \omega_z$ ) =  $2\pi \times (359, 352, 59)$  Hz. Trap frequencies of ( $\omega_{\text{rad}}, \omega_{\text{ax}}$ ) =  $2\pi \times (260, 38)$  Hz are typically obtained from induced BEC trap oscillation measurements. The frequencies are lower than expected due to uncertainties in the trap power estimates, and due to imperfect alignment of the beams. However, the aspect ratio of the trap is similar to what we estimate, and is reproducible over long periods of time<sup>5</sup>. The trap frequencies are measured frequently during measurement campaigns to have them for reference.

# 2.2 Detecting atoms

In our experiments the helium atoms are detected by absorption imaging or by temporal flux detection (which are both destructive measurements). Metastable helium allows for efficient detection on a microchannel plate (MCP) detector due to the large internal energy of 19.82 eV, and we obtain excellent signal-to-noise when measuring the time-of-flight distribution. Unfortunately this method does not allow in-situ or timedelayed observation of the atoms. Hence also absorption imaging is used to observe the atoms.

### MCP detection

If the magnetic trap or optical dipole trap is switched off, the atomic cloud will expand due to its internal energy and momentum distribution in the trap. The metastable atoms expand from the trap center and hit an MCP detector (Hamamatsu F4655, surface radius of 7.25 mm),

<sup>&</sup>lt;sup>4</sup>With a transmission T = 0.9 per window, the power in the trap  $P_{\text{trap}} = P_0(T + T^3) \approx 1.6P_0$ , where  $P_0$  is the power of the first optical dipole trap beam sent into the UHV chamber.

<sup>&</sup>lt;sup>5</sup>Over the course of a few years the alignment of the optical dipole trap of course has had quite some adjustments but we always obtain the same aspect ratio of the trap (7) similar to the value of 6.6 determined by Van Rooij [190].

which is located 17 cm below the trap (see Figure 2.2). The MCP detector is shielded with a metallic grid connected to ground potential to prevent attraction of electrons and ions. When a metastable atom hits a channel on the MCP detector it releases an electron, which is subsequently accelerated over the  $\sim 2$  kV potential to the back of the detector. Through further collisions in the microchannels an avalanche of electrons is created, and the resulting current can be measured at the anode of the detector. In this way the flux of metastable atoms through the surface of the detector can be measured as function of the time-of-flight. The great advantage of an MCP detector is its large gain which - in combination with a pulse discriminator - also allows for single atom or ion detection. Such resolution is experimentally very challenging to achieve with optical techniques.

If a trapped cloud has a size  $\Delta x_0$  and momentum distribution  $\Delta p$ , the size of the expanding cloud after time-of-flight t is  $\Delta x(t) = \Delta x_0 + t$  $(\Delta p/m)t$ . Once the time-of-flight  $t \gg m\Delta x_0/\Delta p$ , the size of the cloud can be effectively described by its momentum distribution and is therefore a clean diagnostic tool to determine the temperature of a cloud. As it turns out, even for the coldest atoms in our experiment the time-offlight of  $t \approx 180$  ms is long enough to be in the regime where the flux of the cloud is solely proportional to the momentum distribution, and this is used to determine the temperature of the thermal atoms and chemical potential of the BEC in a single shot. The advantage of measuring in this regime is that the time-of-flight distributions can be calculated in a straightforward manner and therefore provide analytical fit functions to characterize the gas. A full calculation of the time-of-flight flux signals can be found in Ref. [185], but an overview is given here of the different functions used for the different temperature regimes and the bosonic or fermionic nature of the atoms.

### Thermal distribution

At high temperatures - meaning, temperatures that are significantly larger than the critical or Fermi temperature of the gas - the momentum distribution of the trapped atoms can be described by a classical Maxwell-Boltzmann distribution independent of its quantum statistical behavior. In this limit the time-dependent flux integrated over the surface area of the MCP detector is [185]

$$\Phi_{\rm th}(t) = N_{\rm th} \frac{g}{\sqrt{2\pi}\sigma(t)} \frac{t^2 + t_0^2}{2t} \exp\left(-\frac{x_0^2}{2\sigma(t)^2}\right) \left(1 - \exp\left(-\frac{r_0^2}{2\sigma(t)^2}\right)\right),\tag{2.17}$$

where  $N_{\rm th}$  is the total atom number, g the gravitational acceleration,  $\sigma(t) = t\sqrt{k_B T/m}$  the ballistically expanding size of the cloud,

 $t_0 = \sqrt{2h/g}$  the expected time of arrival of a free falling cloud,  $x_0 = h - gt^2/2$  and  $r_0 = 7.25$  mm the radius of the MCP detector surface. The heigth h = 17 cm is the distance between the trap center and the MCP detector. In this limit the only distinction between <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> is due to the (relatively large) mass difference. The MCP signal cannot provide an absolute number  $N_{\rm th}$  by itself, and therefore has been calibrated [184]. However, for the experiments described here the absolute atom number in the MOT is not relevant, and the MCP can be calibrated in different ways when using a quantum degenerate gas (as will be shown in the next sections). An example of the MCP signal as measured when releasing the cloud from the MOT is shown in Figure 2.3.

#### Ideal Bose and Fermi gas

At ultracold temperatures where the chemical potential of the gas becomes non-negligible compared to the thermal energy of the gas (for our experiments typically around  $T \approx 1 \ \mu\text{K}$ ), the difference between the bosonic and fermionic nature of <sup>4</sup>He<sup>\*</sup> and <sup>3</sup>He<sup>\*</sup> starts playing a role. The Bose-Einstein and Fermi-Dirac statistics need to be taken into account and the 'simple' flux description for a thermal gas no longer holds. In this regime the integrated flux for a Bose gas is [185]

$$\Phi_{\text{Bose}}(t) = \frac{N_{\text{th}}}{g_3(z)} \frac{g}{\sqrt{2\pi\sigma}} \frac{t^2 + t_0^2}{2t} \times \left[ g_{5/2} \left( z \exp\left(-\frac{x_0^2}{2\sigma^2}\right) \right) - g_{5/2} \left( z \exp\left(-\frac{x_0^2 + r_0^2}{2\sigma^2}\right) \right) \right], \quad (2.18)$$

where the functions  $g_n(\varepsilon) = \sum_{k=1}^{\infty} \varepsilon^k / k^n$  are called the Bose functions or polylogarithm functions, and  $z = \exp(\mu/k_B T)$  the fugacity of the gas.



**Figure 2.3** – Time of flight signal on the MCP detector (gray) by releasing atoms from the MOT. Initially the MCP detects background atoms during the MOT loading stage. The loading stops at t = 0 s and after 50 ms the first atoms arrive at the MCP. The signal is fitted using  $\Phi_{th}(t)$  (Eq. 2.17, red line). From the fit we determine T = 0.54 mK and  $N_{th} \approx 3.2 \times 10^8$ .

In the limit where  $z \to 1$   $(k_B T \gg \mu)$ , this flux expression reduces to the classical Maxwell-Boltzmann flux Eq. 2.17.

For the Fermi gas the integrated flux signal is almost the same as in Eq. 2.18 apart from the substitution  $g_n(\varepsilon) \to -g_n(-\varepsilon)$ , resulting from the change of a Bose-Einstein distribution to a Fermi-Dirac distribution. Contrary to the Bose gas which has a phase transition at its critical temperature, the Fermi gas has a smooth and continuous transition into the quantum degenerate regime. In a harmonic oscillator trap with radial and axial trap frequencies  $\omega_{\rm rad}$  and  $\omega_{\rm ax}$ , geometric average trap frequency  $\bar{\omega} = (\omega_{\rm rad}^2 \omega_{\rm ax})^{1/3}$ , and containing N identical fermions, the Fermi temperature  $T_F$  is [200, 201]

$$k_B T_F = \hbar \bar{\omega} (6N)^{1/3}.$$
 (2.19)

To illustrate, our typical trap frequencies  $\omega_{\rm rad} = 2\pi \times 260$  Hz and  $\omega_{\rm ax} = 2\pi \times 40$  Hz result in  $T_F \approx 1.2 \,\mu{\rm K}$  for  $10^6$  atoms and  $T_F \approx 0.56 \,\mu{\rm K}$ 

for  $10^5$  atoms. As we are able to cool about  $10^5 - 10^6$  <sup>3</sup>He<sup>\*</sup> atoms to  $T \approx 0.2 \ \mu\text{K}$  it is clear that degenerate Fermi gases in the regime  $T/T_F \approx 0.2 - 0.4$  can be produced, which is definitely cold enough to observe deviations in the time-of-flight distribution from the Maxwell-Boltzmann distribution. Furthermore, in this regime the chemical potential of the Fermi gas can be estimated using the Sommerfeld expansion [200]

$$\mu = k_B T_F \left[ 1 - \frac{\pi^2}{3} \left( \frac{T}{T_F} \right)^2 \right], \qquad (2.20)$$

which implicitly depends on the total number of atoms as shown in Eq. 2.19. By determining T and  $\mu$  of the Fermi gas from the MCP time-of-flight distribution and inverting Eqns. 2.19 and 2.20, one can determine the total atom number N in the cloud. This approach provides a self-consistent cross-check with the calibration done using the Bose-Einstein condensate, as used in Ref. [156] and explained in the next section.

#### **Bose-Einstein condensate**

Whereas fermions gradually enter the quantum degenerate regime when cooling down, bosons exhibit dramatic different behavior due to the different quantum statistics. The occupancy of the lowest state increases monotonously as a cloud of N trapped identical bosons cools down until the occupation of the lowest state becomes equal to the sum over all other states. This condition is parametrized by the critical temperature  $T_c$  [202]

$$k_B T_c = 0.94 \hbar \bar{\omega} N^{1/3}, \qquad (2.21)$$

where  $N = N_{\text{BEC}} + N_{\text{th}}$  is the sum of the atoms in the Bose-Einstein condensate and the thermal fraction of the gas. Below the critical temperature  $T_c$  the occupancy of the Bose-Einstein condensate in a harmonic trap scales as [202]

$$\frac{N_{\rm BEC}}{N} = 1 - \left(\frac{T}{T_c}\right)^3. \tag{2.22}$$

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Combining Eqs. 2.21 and 2.22 using the same typical trap parameters as for the Fermi gas and realizing that we produce Bose-Einstein condensates with ~  $1 \times 10^6$  atoms in the condensed fraction, the critical temperature is around  $T_c \approx 0.6 - 0.8 \ \mu\text{K}$ . With a typical temperature  $T \approx 0.2 \ \mu\text{K}$  of the thermal fraction and using Eq. 2.22, this is in agreement with the ratio of  $N_{\text{BEC}}/N \approx 0.97 - 0.99$  generally observed in the optical dipole trap.

The wavefunction  $\Psi_0(\vec{r}, t)$  of a Bose-Einstein condensate trapped in a potential  $V_{\text{ext}}(\vec{r}, t)$  is described by the Gross-Pitaevskii equation [202]

$$\left(-\frac{\hbar^2 \nabla^2}{2m} + V_{\text{ext}}(\vec{r},t) + \tilde{g} |\Psi_0(\vec{r},t)|^2\right) \Psi_0(\vec{r},t) = \mu \Psi_0(\vec{r},t), \quad (2.23)$$

where  $\tilde{g} = 4\pi\hbar^2 a/m$  is the coupling constant of the interaction term determined by the the s-wave scattering length a between the atoms. The limiting case where the kinetic energy of the Bose-Einstein condensate can be neglected with respect to the interaction energy, is called the Thomas-Fermi approximation. The validity of the Thomas-Fermi approximation can be estimated by considering the Thomas-Fermi parameter  $N_{\rm BEC}(a/a_{\rm ho})$  [202] using the measured s-wave scattering length a = 7.512(5) nm [152], the harmonic trap oscillator length defined as  $a_{\rm ho} = \sqrt{\hbar/m\bar{\omega}} \approx 7.5 \ \mu m$ , and  $N_{\rm BEC} \approx 10^6$ . We estimate that  $N_{\rm BEC}(a/a_{\rm ho}) \approx 10^3 \gg 1$ , indicating that the Thomas-Fermi approximation is valid (and only starts to break down for a condensate with  $N_{\rm BEC} \approx 10^3$ ).

In the Thomas-Fermi limit the solution of the Gross-Pitaevskii equation simplifies to the wave function  $\Psi_{\rm TF}(\vec{r},t) = \sqrt{n_{\rm TF}(\vec{r},t)}$ , with

$$n_{\rm TF}(\vec{r},t) = \frac{1}{\tilde{g}} [\mu(t) - V_{\rm ext}(\vec{r},t)], \qquad (2.24)$$

being the density distribution of the Bose-Einstein condensate in the trap potential  $V_{\text{ext}}(\vec{r},t)$ . For a harmonic potential the density distribution is the well-known inverted parabola with spatial cutoff points where the chemical potential is equal to the trap potential. Applying the normalization condition for the total atom number  $N_{\text{BEC}}$  gives the Thomas-Fermi relation between the atom number and chemical potential.

tial of the condensate

$$\mu = \frac{1}{2} \left( 15a\bar{\omega}^3 \hbar^2 m^{1/2} \right)^{2/5} N_{\text{BEC}}^{2/5}.$$
 (2.25)

When a Bose-Einstein condensate is released from a trap the internal (mean field) energy is converted in kinetic energy and determines the momentum distribution of the cloud<sup>6</sup>. As the mean field energy is determined by the density distribution, the momentum distribution of the expanding cloud is described by a rescaled parabolic shape related to the trapped density distribution of the condensate. If the total condensate falls on the MCP detector surface, the flux is [185]

$$\Phi_{\rm BEC}(t) = N_{\rm BEC} \frac{15}{16} g \frac{t^2 + t_0^2}{2t^2} \sqrt{\frac{m}{2\mu}} \max\left[0, 1 - \frac{m}{2\mu} \left(g \frac{t^2 - t_0^2}{2t}\right)^2\right]^2.$$
(2.26)

In general the MCP detects a thermal fraction with a Bose-Einstein condensate superposed, in which case a fit function  $\Phi(t) = \Phi_{\text{BEC}}(t) + \Phi_{\text{Bose}}(t)$  is used to determine all parameters simultaneously. A typical MCP signal measured with a Bose-Einstein condensate released from the optical dipole trap is shown in Figure 2.4.

Only for the largest Bose-Einstein condensates that we can make, the mean field energy would be large enough for the cloud to expand to a larger size than the MCP surface, and this would lead to a small correction of the chemical potential. As the chemical potential can be related to the number of atoms in the condensate, Eq. 2.25 can be combined with time-of-flight measurements of the Bose-Einstein condensate to provide an absolute calibration of the MCP detector, as was done in Ref. [156]. The advantage of this calibration is that the chemical potential is related to the temporal width of the time-of-flight signal and not to the absolute magnitude, thereby being less sensitive to saturation effects of the MCP detector. This calibration is still used in the current measurements, and is found to be in agreement with the alternative calibration using the Fermi gas as discussed in the previous section.

<sup>&</sup>lt;sup>6</sup>The momentum distribution of a trapped Bose-Einstein condensate therefore looks quite different than the momentum distribution of a released condensate [203].



**Figure 2.4** – Time of flight signal on the MCP detector (gray) by releasing a Bose-Einstein condensate from the optical dipole trap. The signal is fitted using the bimodal distribution  $\Phi(t) = \Phi_{BEC}(t) + \Phi_{Bose}(t)$ (Eqs. 2.18 and 2.26, red line). The individual time of flight signals are shown for the BEC (black line) and the thermal fraction (black line, dashed). The chemical potential of the BEC  $\mu/k_B \approx 0.5 \ \mu K$  and the temperature of the thermal fraction  $T \approx 0.4 \ \mu K$ . The atom numbers are  $N_{BEC} \approx 6 \times 10^6$  and  $N_{th} \approx 1 \times 10^5$ .

When measuring the time-of-flight signal of a cloud released from the magnetic trap the residual magnetic fields can apply an additional force on the atoms. This can alter the momentum distribution and this, in combination with large condensates, is observed in Ref. [125]. The so-called 'pushing' of the cloud by transient magnetic fields (which can also play a role for the thermal flux signals from a MOT) can be parametrized by substituting  $t \to t + \Delta t$ , where  $\Delta t$  is a free fit parameter for the timing offset. This problem is not present when releasing the condensate from an optical dipole trap.

### Ion MCP detection

A second, unshielded, MCP detector is placed approximately 11 cm above the trap center and off-centered to prevent blocking of beams. Applying a negative voltage to the front end of the MCP detector will attract He<sup>+</sup> ions created by the Penning ionization process as discussed in Section 2.1. The ion signal provides a probe of the density of the trapped cloud as the ion production rate scales nonlinear with the density. This signal has been used in previous experiments to measure ionizing collision rates [148, 149, 153, 180] or for photoassociation spectroscopy [150, 181]. It was also used to observe the onset of Bose-Einstein condensation, as the sudden increase in density leads to a sudden increase in ion production due to two- and three-body collisions [125].

In the current experiments the ion MCP is incidentally used for diagnostic purposes, but not for quantitative analysis. It was last used to observe the  $2 {}^{3}S_{1} - 2 {}^{1}P_{1}$  transition at 887 nm for the first time by scanning the spectroscopy laser wavelength in a continuously loading MOT. The increased on-resonance scattering losses would lead to a decrease in MOT density and therefore lower ion production rate.

### Absorption imaging

Although the MCP detector has excellent signal-to-noise ratio and gain, it is placed at a fixed distance with respect to the trap center and cannot explore the time-dependent behavior of the expanding cloud for various expansion times. Therefore we also use the well-known absorption imaging technique, which is a versatile tool used in almost any cold atom experiment.

Absorption imaging is a technique which essentially measures the 'shadow' of a cloud of atoms on a CCD camera due to the absorption of resonant light by the atoms. If the trapping potential is well known, the size and optical thickness of the shadow can be related to the temperature and atom number of the cloud. Furthermore, absorption imaging can be used in a time-delayed configuration to observe dynamical effects in the cloud, from simple atom loss to trap oscillations or density fluctuations. In our experiment we use a Xenics Xeva-1.7-320 camera, which consists of a  $320 \times 256$  array of  $30 \times 30 \ \mu\text{m}^2$  pixels. The camera is InGaAs

based, and was chosen in the last few years over a silicium-based CCD camera (Hamamatsu C4880) for its large quantum efficiency (80%) with respect to the silicium-based quantum efficiency of < 1.5%. Although the Xenics camera has a larger noise floor than the Hamamatsu camera, its signal-to-noise is five times larger and therefore preferred.

For low intensities  $I \ll I_{\text{sat}}$  (see Table 2.1 in Section 2.3), typically  $I \approx I_{\text{sat}}/10$ , the transmitted intensity of a probe beam decreases exponentially with the optical thickness of the cloud as

$$I_{\rm out}(x,y) = I_{\rm in}(x,y)e^{-\sigma_{\rm abs}n(x,y)},$$
(2.27)

where  $\sigma_{abs}n(x, y)$  is the optical thickness, and n(x, y) the column density integrated along the direction of propagation of the probe beam (which contains the temperature and absolute atom number of the trapped cloud). The absorption cross section  $\sigma_{\rm abs} = \hbar \omega_0 (\Gamma/2) / I'_{\rm sat}$ , with  $\omega_0$ the transition frequency,  $\Gamma/2\pi$  the linewidth and  $I'_{\rm sat} = fI_{\rm sat}/\chi$  the effective saturation intensity. The saturation intensity is modified by a factor f due to the exact population of the magnetic sublevels and the polarization of the light (f = 17/10 for spin-stretched atoms and)linear polarized light, but f = 18/10 for unpolarized atoms). The temperature-dependent line shape factor  $\chi$  is based on a convolution of Maxwell-Boltzmann velocity distribution of the atoms and the absorption lineshape. Both f and  $\chi$  are discussed extensively by Tol [184]. The column density n(x, y) can be determined by measurement of  $I_{in}(x, y)$ and  $I_{out}(x, y)$  combined with an accurate calculation of the absorption cross section. This is done by consecutive acquisition of three images on the CCD camera. The first image  $I_{abs}$  is taken with the probe beam and the atoms. After waiting for 300 ms another image  $I_{\text{probe}}$  is taken solely with the probe as the atoms are gone. The third image is taken (again with a 300 ms interval) without the probe light, thereby providing a background  $I_{\rm bgr}$ . The normalized transmission can then be computed as

$$\frac{I_{\rm out}(x,y)}{I_{\rm in}(x,y)} = \frac{I_{\rm abs}(x,y) - I_{\rm bgr}(x,y)}{I_{\rm probe}(x,y) - I_{\rm bgr}(x,y)}.$$
(2.28)

A recent addition to this computation was the so-called 'fringe removal algorithm', which removes artefacts in the normalized image caused by variations in the intensity of the probe light between  $I_{\rm abs}$  and  $I_{\rm probe}$  [190]. This post-processing algorithm improves the quality of the images significantly. For a 1-to-1 imaging system as used here, the normalized transmission can be calculated on a pixel-by-pixel basis and directly correlated to the column-integrated spatial distribution of the cloud by

$$n(x,y) = -\frac{1}{\sigma_{\rm abs}} \ln\left(\frac{I_{\rm out}(x,y)}{I_{\rm in}(x,y)}\right). \tag{2.29}$$

Depending on the the isotope and temperature, this distribution can be fitted with an appropriate function to extract the physical parameters, just as for the MCP time-of-flight signals.

#### **Imaging fit functions**

A full derivation of the imaging fit functions is given in Ref. [185], and only the main results are repeated here for reference. For a thermal distribution of atoms in a harmonic trap, the column-integrated density distribution is

$$n_{\rm th}(x,y) = \frac{N_{\rm th}}{2\pi\sigma_x\sigma_y} \exp\left(-\frac{x^2}{2\sigma_x^2} - \frac{y^2}{2\sigma_y^2}\right),\tag{2.30}$$

where  $N_{\rm th}$  is the total atom number and  $\sigma_i$  the width of the expanding cloud (i = x, y are the two orthogonal axes in the imaging plane). The width of the expanding cloud along axis *i* scales with the trapping frequency as  $\sigma_i(t) = \sigma_i(0)\sqrt{1 + \omega_i^2 t^2}$ . Assuming thermal equilibrium and equipartition of energy, the initial size of the trapped cloud is  $\sigma_i(0) = \sqrt{k_B T / m \omega_i^2}$ . For  $t \gg \omega_i^{-1}$  the size of the cloud asymptotically increases as  $t\sqrt{k_B T / m}$  along every axis. This shows that one can use the time-dependent expansion of the cloud to do thermometry, but also that information about the initial density distribution is lost for a ballistically expanding cloud.

Once the temperature of the cloud is near to the critical temperature or the Fermi temperature, quantum statistics starts playing a role in the density distributions similar to the MCP flux distributions. The appropriate fit function becomes [185]

$$n_{\text{Bose}}(x,y) = \frac{N_{\text{th}}}{2\pi\sigma_x\sigma_y g_3(z)} g_2\left(z\exp\left(-\frac{x^2}{2\sigma_x^2} - \frac{y^2}{2\sigma_y^2}\right)\right),\qquad(2.31)$$

with again  $z = \exp(\mu/k_B T)$  the fugacity and  $g_n(\varepsilon)$  the Bose functions as discussed before. The density distribution of a Fermi gas is obtained by the substitution  $g_n(\varepsilon) \to -g_n(-\varepsilon)$ .

Below  $T_c$  the density distribution of the Bose-Einstein condensate can be described in the Thomas-Fermi limit, and the parabolic density distribution appears as

$$n_{\rm BEC}(x,y) = \frac{5}{2\pi} \frac{N_{\rm BEC}}{r_x r_y} \max\left[0, 1 - \left(\frac{x}{r_x}\right)^2 - \left(\frac{y}{r_y}\right)^2\right], \qquad (2.32)$$

with  $r_i$  (i = x, y) the radius of the trapped or expanding condensate. If the condensate is trapped, the radius is  $r_i = \sqrt{2\mu/m\omega_i^2}$  and known as the Thomas-Fermi radius [202]. When the condensate expands there can be a large difference between the x and y direction if they have very different trapping frequencies (i.e. they represent the axial and radial directions, respectively). For an aspect ratio  $\epsilon = \omega_{\rm ax}/\omega_{\rm rad} \ll 1$ , and a dimensionless expansion time  $\tau = \omega_{\rm rad} t$  the radii increase as [204]

$$r_{\rm rad}(\tau) = r_{\rm rad}(0)\sqrt{1+\tau^2},$$
 (2.33)

$$r_{\rm ax}(\tau) = r_{\rm ax}(0) \left[ 1 + \epsilon^2 \left( \tau \arctan(\tau) - \ln \sqrt{1 + \tau^2} \right) \right]. \tag{2.34}$$

This is radically different compared to the expansion behavior of the other (non-condensed) gases. The reason is the large interaction energy in the condensate, which allows the Gross-Pitaevskii equation to be rewritten in terms of hydrodynamic equations of a superfluid [202]. In this regime the hydrodynamic expansion in the orthogonal directions is not independent as for the thermal gases, leading to the modified time-dependent behavior as shown here. As a result of the hydrodynamic behavior the aspect ratio  $\epsilon$  of the expanding condensate actually inverts to  $2/\pi\epsilon$  as cloud expands faster along the high trap frequency (radial) axis than along the axial direction; this is generally seen as a smoking gun for the presence of a condensate.

Similar to the MCP fit functions for a thermal Bose cloud and a condensate, the absorption image can be fit by a combined function  $n_{\rm th}(x, y) + n_{\rm BEC}(x, y)$  to extract information about the thermal atoms and the condensate simultaneously.

## Magnetic field measurements

With the implementation of the optical dipole trap, rf-induced transitions (as mentioned in Section 2.1 to do forced evaporative cooling in the magnetic trap) became available as a metrology tool to determine the local magnetic field, as shown by Van Rooij [190]. This is done in a Stern-Gerlach type of measurement [205], where a short (typically  $\sim 40 \ \mu s$ ) burst of rf radiation transfers part of the He<sup>\*</sup> atoms from the spin-stretched  $m_I = +1$  ( $m_F = +3/2$  for <sup>3</sup>He<sup>\*</sup>) state to the other magnetic substates, effectively creating a spin-mixture. The atoms are immediately released from the optical dipole trap, and a magnetic field gradient is switched on simultaneously to spatially separate the different magnetic substates. After a time-of-flight of  $\sim 10$  ms the atoms are imaged using the absorption imaging setup, and the population of the different spin states can be spatially resolved. If the spin-dependent losses are negligible (which is the motivation for releasing the atoms immediately after the rf burst), the relative population of the different spin states, also known as the 'spin-flip ratio' can be measured as function of the frequency of the applied rf field. The magnetic field can be determined from the resonance frequency at which the largest spin-slip ratio is achieved, combined with the theoretical known Zeeman shift of the states as given in Section 2.1.

The magnetic field- or Zeeman shift, measurements were essential in the  $2 {}^{3}S - 2 {}^{1}S$  frequency metrology to determine the systematic frequency shift caused by the background magnetic field [31]. Sub-kHz (typically 0.5 kHz) accuracy could be achieved for a single magnetic field measurement (i.e. a resonance scan), and repetition of the measurement throughout a single day revealed a Gaussian distribution of the determined Zeeman shift (magnetic field) with a standard deviation of 2 kHz [190]. For the future sub-kHz accuracy goal on the  $2 {}^{3}S - 2 {}^{1}S$  isotope shift, the Zeeman shift needs to be measured with better accuracy than currently available and should be better understood. First results

of a new method, based on a Ramsey-type measurement [206], will be discussed in Chapter 5.

# 2.3 Optical setup

The previous sections deal with the general operation of the experimental setup and the detection methods. The optical and laser systems that are currently used to generate all the required light will be discussed in this section. The new narrow linewidth laser system used for the 2  ${}^{3}S_{1} - 2 {}^{1}S_{0}$  spectroscopy work will be separately discussed in Section 2.5.

## Lasers for trapping, cooling, and imaging He\*

The laser systems had a big revision by Van Rooij [190], who introduced the ytterbium-doped fiber laser system (Koheras Adjustik Y10 by NKT Photonics) for laser cooling and trapping purposes. The advantage of the fiber laser system compared to the old diode laser systems is the narrow linewidth, relatively large power availability, and day-to-day stability of the system. Therefore the multiple laser systems were replaced by a single Koheras fiber laser combined with a Keopsys amplifier for  ${}^{4}\text{He}^{*}$ and a Toptica DL100 diode laser combined with a vtterbium-doped fiber amplifier (NuAMP by Nufern) for <sup>3</sup>He<sup>\*</sup>. We have made this configuration even more robust by replacing the Keopsys amplifier with another Nufern amplifier and the Toptica laser with a Koheras Adjustik fiber laser. The wavelength of the fiber laser systems can be temperature tuned with a responsivity of 6 GHz/K over a range of  $\sim 200$  GHz, and allow fast modulation (bandwidth  $\sim 20$  kHz) with a piezo transducer (0-200 V) over a range of 4 GHz. The Nufern amplifiers can both amplify to slightly over 2 W, but are generally used to provide about 700 mW for the experiment. The two identical Koheras Adjustik/Nufern amplifier configurations for <sup>4</sup>He<sup>\*</sup> and <sup>3</sup>He<sup>\*</sup> have the (unfortunately already proven) advantage of flexibility of configuration in case of malfunction of a single system.

For laser cooling, trapping, and imaging applications the  $2 {}^{3}S_{1} - 2 {}^{3}P_{2}$ (D2) transition is used for <sup>4</sup>He<sup>\*</sup> and the  $2 {}^{3}S_{1}(F = 3/2) - 2 {}^{3}P_{2}(F = 5/2)$ (C3) transition for <sup>3</sup>He<sup>\*</sup>, of which the relevant properties are shown in

**Table 2.1** – Relevant atomic and laser cooling properties of <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup>. The transition-specific parameters are calculated using the following equations:  $I_{sat} = \pi \hbar \Gamma/3\lambda^3$ ,  $k_B T_{Dop} = \hbar \Gamma/2$ ,  $k_B T_{rec} = \hbar^2 k^2/2m$  with  $k = 2\pi/\lambda$ , and  $v_{rec} = \hbar k/m$ . The linewidths  $\Gamma/2\pi$  differ at the 10<sup>-4</sup> level due to the <sup>3</sup>He-<sup>4</sup>He mass difference, which is neglected here.

Atomic property	$^{4}\mathrm{He}^{*}$	${}^{3}\mathrm{He}^{*}~(F=3/2)$	Ref.
Mass [amu]	4.00260325413(6)	3.0160293201(25)	[88]
Natural abundance	$\geq 0.999959$	$\leq 0.000041$	[207]
Lifetime [s]	$7.9(5) \times 10^{3}$	$7.9(5) \times 10^{3}$	[59]
Nuclear spin	0	1/2	
Laser cooling			
Cycling transition	$2 \ {}^3S_1$ -2 $ {}^3P_2$	$2 {}^{3}S_{1} (F = 3/2)$ -	
		$2^{3}P_{2} (F = 5/2)$	
Wavelength [nm]	1083.33064	1083.46246	[30]
$\Gamma/2\pi$ [MHz]	1.62	1.62	[56]
$I_{\rm sat} \ [{\rm mW/cm^2}]$	0.167	0.167	
$T_{\rm Dop} \ [\mu {\rm K}]$	38.4	38.4	
$T_{\rm rec} \; [\mu {\rm K}]$	2.04	2.72	
$v_{\rm rec}   [{\rm m/s}]$	0.092	0.123	

Table 2.1. These transition frequencies are separated by ~ 35 GHz, which cannot easily be covered by a single laser system with (e.g. EOM-generated) sidebands. Therefore both lasers individually frequency stabilized in an identical way, using identical electronics and two saturated absorption spectroscopy setups (see, e.g. Refs. [186, 187]) consisting of an rf discharge cell filled with either <sup>3</sup>He or <sup>4</sup>He to populate the <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> states. The laser frequency is modulated at a frequency of 2 kHz using a lock-in amplifier system (EG&G 5209). The Lamb dip signal (i.e. the saturated absorption peak in the center of a Doppler broadened absorption dip) measured with a photodiode is demodulated by the lock-in amplifier system to generate a dispersive 'error' signal. The error signal is fed into a VU-designed digital PID 'lock box' to provide a (0-10 V) feedback signal. This feedback signal is linearly amplified to 0-140 V by a VU-designed voltage amplifier and sent to the piezo transducer of the

fiber laser. Manual control over the feedback voltage allows scanning of the laser frequency when looking for the dispersion signal. The lock-in amplifier settings for both the <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> systems are different due to different signal amplitudes and phase delays, but the PID settings are quite similar as the laser systems are identical. The fiber lasers are truly turnkey systems and have excellent day-to-day stability and reproducibility (they rarely require adjustment of parameters). Relocking to the transition frequency is only required in strenuous circumstances (i.e. if there are significant temperature changes in the laboratory).

## Optical dipole trap

The optical dipole trap is operated by a 1557-nm Scorpio laser system from NP Photonics containing an erbium-doped seed and amplifier module with a total output of over 2 W. This laser system was simultaneously used for trapping and spectroscopy of the  $2^{3}S - 2^{1}S$  transition [31], but we have switched to a different laser system for spectroscopy (see Section 2.5). The Scorpio laser is connected to the laboratory computer using an RS-232 connection for controlling the wavelength and output power, and for monitoring the performance of the laser system. Currently the laser is used in a free running mode at a wavelength of 1557.3 nm, and part of the light (~ 10 mW) is picked off by the He\*Rb experiment to seed an erbium-doped Nufern amplifier (10 W) for their own optical dipole trap.

For long-term power stabilization of the optical dipole trap power, a motorized rotation stage (Thorlabs PRM1/MZ8 + TDC001 controller) is implemented and mounted with a  $\lambda/2$  waveplate in combination with a polarizing beam splitter to allow digital control over the optical dipole trap power. The power is monitored by a photodiode (Thorlabs PDA255), and digital acquisition (Agilent 34401A digital multimeter) of the signal allows for small corrections to the optical dipole trap power per single measurement. In the line shape measurements which take over one hour to acquire (see Chapter 4), this greatly improved the stability of the signals.

### Optics and laser beams

Apart from the laser systems, another overhaul by Van Rooij was to implement single-mode polarization maintaining optical fibers in the setup [190] to decouple the Zeeman slower, MOT and imaging beams between the optical table and the cold atom setup. Only the beams of the collimation section, spin polarization and 1D Doppler cooling are still free-space, but this does not seem to be negatively affecting the stability of the experiment. Furthermore, the optical paths of the <sup>3</sup>He<sup>\*</sup> and <sup>4</sup>He<sup>\*</sup> light are now overlapped in a robust way using a 2 × 2 fused fiber coupler. In this way the need to realign all optical beams is reduced significantly by using one output of the fused fiber coupler for the collimation section and the other output for all other applications. The initial optical power requirement is currently ~ 700 mW due to insertion losses of the fibers and acousto-optical modulators (AOMs), which is generally 20-30%. This can easily be covered with the current Nufern amplifier systems (which can go up to 2 W).

The various beams need to be at different detunings with respect to the absolute transition frequency, and various AOMs are used for this purpose. The optical power of each beam can be tweaked to its specific need by adjusting the rf power of the AOM or by redistributing the light with a half-wave plate and a polarizing beam splitter. The design and physical layout has not changed with respect to the previous thesis [190], and only a schematic overview of the optical setup is shown in Figure 2.5 with all relevant beam powers and detunings.



**Figure 2.5** – Schematic overview of the optical system. The separate  ${}^{3}He^{*}$  and  ${}^{4}He^{*}$  laser systems are locked individually to the respective laser cooling transitions, and both seed a Nufern fiber amplifier with a nominal free-space output power of ~ 700 mW. Both beams are coupled into a 50 : 50 fiber splitter to guarantee spatial overlap, and one optical path is fully dedicated to the collimation section. The other path is split and sent through various AOMs (detunings indicated inside the boxes) to generate the required detuning. The right-hand side indicates the purpose of each beam and the optical power as used per isotope at the experiment. The  ${}^{4}He^{*}$  laser system additionally seeds amplifier systems for two other  ${}^{4}He^{*}$ -based experiments in our laboratory.

# 2.4 Digital control of the experiment

The LabView-based control software of the experiment was replaced by a Python based software control and user interface by Van Rooij [190]. The Python software provides more flexibility when controlling different devices and interfaces, and was therefore preferred over the old system. Due to the modular design of the software it has been relatively straightforward to add devices and their appropriate control and configuration commands. The digital and analog control of the experiment ran with a 64-channel digital card (Viewpoint Systems DIO-64) and a 8-channel analog card (National Instruments PCI-6713). A timetable of a single experimental run is programmed to the digital card, and runs independent of the clock speed of the computer<sup>7</sup>. Simultaneously a timetable is programmed to the analog card, and it runs synchronously with the digital card (imposed by TTL triggers from the digital card). The output channels of the digital card are 5 V TTL triggers, and the output of the analog card spans a  $\pm 10$  V range with  $\sim 5$  mV resolution (12-bit). The digital card has a clock frequency of 50 MHz, enabling a maximum speed of 25 MHz (i.e. a timing resolution 40 ns). The rise/fall time of the TTL pulses is  $\sim 50 \ \mu s$ , which means that we are able to configure TTL pulses with  $< 1 \ \mu s$  resolution but the minimum pulse duration is  $\sim 100 \ \mu s$ . This is no limiting factor for our experiments, but the timing resolution was initially used for our Rabi and Ramsey-type measurements with rf pulses (see Chapter 5) until we switched to an arbitrary waveform generator.

After a breakdown of the main experiment computer in January 2016 (with the obsolete Windows XP operating system), the DIO-64 Viewport Systems digital card could no longer be supported by the new operating system (Windows 7) and the whole interface was upgraded to two 32-channel digital cards (National Instruments PCIe-6536B digital I/O card with SMB-2163 digital I/O rack) and one 8-channel analog card (National Instruments PCI-6733). This also required an upgrade of the back-end control software (which was written in C++) to Python

<sup>&</sup>lt;sup>7</sup>This configuration prevents any unwanted buffering or delays caused by background software of the computer itself. The timetable is uploaded to the digital card at the beginning over each measurement, and the computer is idle while the sequence runs on the digital card.

code and could be implemented based on code further developed by the other He<sup>\*</sup> experiments, who are also using the Python program. The new digital cards have a clock frequency of 25 MHz, but this reduction of a factor of two in the timing resolution poses no practical limitations for the experiment. Also, the new TTL pulse height is only 3.3 V compared to the former 5 V but no issues were discovered after implementing the new TTL signals. The new analog card has the same  $\pm 10$  V range as the old card but better resolution (2.24 mV, 13-bit). A ground loop in the new analog board caused the current control of the magnetic trap to display cross-talk between the different current supplies, which was solved by isolating the analog signals using Delta Elektronica ISO AMP modules to provide a floating voltage.

Apart from the digital and analog control, the experiment control software also configures devices and reads out data at the end of the measurement through USB, RS-232, or GPIB (i.e. the MCP time-of-flight signal from an oscilloscope). This is all parallel to the experimental sequence running from the digital and analog boards. Specific unavoidable computer-controlled actions which are timing-sensitive with respect to the digital board sequence<sup>8</sup> are timed by monitoring dedicated TTL channels from the digital card with a separate National Instruments DAC module. The complete timetable, filenames, and settings of devices are all stored in a single script file per single measurement, such that all data acquired from various devices is fully traceable for (automated) analysis afterwards.

The Xenics camera is initiated and controlled via a separate program alongside the main program, but it can be controlled via the main program user interface. Data acquisition and processing occurs through the separate program, but the main program is able to access the files and generate the absorption images for analysis.

During the original  $2 {}^{3}S - 2 {}^{1}S$  measurement [31] the NP Photonics laser was digitally frequency stabilized to a femtosecond frequency comb. The infrastructure existed of a fiber link to the frequency comb laboratory, a free-space optical system to create a beatnote between the NP Photonics laser light and the frequency comb on an avalanche photodiode

<sup>&</sup>lt;sup>8</sup>For example, a controlled ramp of the optical dipole trap power by rotating the Thorlabs rotation stage during the optical dipole trap loading stage.

detector, and a frequency counter to measure the beatnote. The measured beatnote frequency was transferred via an ethernet connection to the experiment computer, where a continuous PID feedback signal was calculated and sent to the NP Photonics laser using a National Instruments DAC module. The continuous ethernet connection was mediated by a secondary computer using a Linux-based operating system in order minimize interference with the experimental control sequence on the Windows computer. This infrastructure was also used in the  $2 \ ^3S_1 - 2 \ ^1P_1$  experiment to frequency stabilize the Coherent 899-21 Ti:sapphire ring laser (see Chapter 3), but for the latest work on the  $2 \ ^3S - 2 \ ^1S$  transition (Chapter 4) it has become obsolete as the frequency stabilization scheme is completely different to achieve a much smaller linewidth of the spectroscopy laser. This will be discussed in the next section.

# 2.5 A new spectroscopy laser system

In the first observation and measurement of the 2  ${}^{3}S - 2 {}^{1}S$  transition by Van Rooij [31, 190], the spectroscopy laser was digitally frequency stabilized to the frequency comb (see Sections 2.3 and 2.4). The 30 ms integration time of the frequency counter used in this configuration limited the bandwidth of the feedback loop to 35 Hz. As a result of this bandwidth the linewidth of the spectroscopy laser at the ~ 1 s timescale of the measurement phase was on the order of 80 – 100 kHz as observed in the linewidth of the absorption spectra. A histogram of the frequency deviations of the spectroscopy laser with respect to the frequency comb at the 30 ms timescale showed a Gaussian FWHM of 67 kHz [190], which can be roughly interpreted as a lower limit of the stability of the frequency-stabilized spectroscopy laser.

As a result of this broad linewidth no asymmetry in the line shapes for both  ${}^{4}\text{He}{}^{*}$  and  ${}^{3}\text{He}{}^{*}$  was observed although this could be expected. Without any additional information the absorption features were analysed using Gaussian line shape functions. In this case any nonlinear effects would appear as a nonlinear shift in the determined transition frequency. For  ${}^{4}\text{He}{}^{*}$  no nonlinearities in the ac Stark shift extrapolations was observed, nor an apparent density-dependent (mean field) shift. For the <sup>3</sup>He<sup>\*</sup> spectra, saturation effects seemed to occur in the ac Stark shift extrapolation, which had to be taken care of by ensuring full thermalization of the Fermi gas in the optical dipole trap [190]. Even with all this work, the ac Stark shift extrapolation uncertainty of 1.4 kHz was the limiting factor in the accuracy of the transition frequency, followed by the accuracy of the mean field shift determination for <sup>4</sup>He<sup>\*</sup>.

In order to make the next leap to sub-kHz absolute accuracy, the linewidth of the spectroscopy laser has to be reduced significantly for two reasons. First, it will simply improve the statistical accuracy in determining the absolute transition frequency. Second, a sufficiently narrow spectroscopy laser will reveal any asymmetry in the line shape of the spectrum caused by any underlying physics. A back-of-the-envelope calculation of the relevant broadening scales for the line shapes of both <sup>3</sup>He<sup>\*</sup> (given by the Fermi temperature ~ 40 kHz) and <sup>4</sup>He<sup>\*</sup> (mean field shift of the <sup>4</sup>He<sup>\*</sup> state ~ 10 kHz) suggests that a spectroscopy laser linewidth of < 10 kHz would be sufficient to observe any asymmetries in the line shapes. Of course there are other solutions as well and these are proceeding in parallel to the improvement of the spectroscopy laser linewidth, as will be discussed in Chapter 5.

Parts of the improvements and used infrastructure mentioned in this section are developed and maintained in collaboration with the Ultrafast Laser Physics and Precision Metrology group of Kjeld Eikema.

### Frequency comb for precision metrology

The problem with high-precision metrology of optical transition frequencies  $(10^{14} - 10^{15} \text{ Hz})$  is that they are difficult to measure using conventional electronic techniques. From a metrology point of view the problem also lies with the fact that the definition of the SI second is in the microwave frequency regime, defined by the 9 192 631 770 Hz hyperfine transition of the caesium-133 atom [208, 209], especially as it is typically mediated in the radio frequency regime by a standardized 10 MHz or 100 MHz reference oscillator frequency. The over six orders of magnitude difference between the SI second frequency reference and the optical frequencies poses a serious problem in measuring absolute optical transition frequencies. Initially this was solved by a phase-coherent chain of lasers, starting from the far infrared range ( $\lambda \approx 100 \ \mu\text{m}$ ) down



**Figure 2.6** – The schematic output of a frequency comb, both in the time and frequency domain. A continuous train of pulses separated by time T and with a fixed phase relationship governed by a phase  $\varphi_{ceo}$  can be described in the frequency domain as a regular comb-like mode structure with mode separation  $f_{rep} = 1/T$  and an overall offset frequency  $f_{ceo}$ . The frequency of the n-th mode is given as  $f_n = f_{ceo} + n \times f_{rep}$ .

to the visible range ( $\lambda \approx 650$  nm) [210], but this was far from a practical situation as it required multiple lasers being phase-locked for an extended amount of time.

At the turn of the century a solution was presented using femtosecond pulse mode-locked lasers now known as frequency combs [211–214]. By generating a continuous train of phase-coherent pulses separated by a time T within a cavity, the Fourier spectrum of the light portrays a regular (comb-like) mode structure with frequency spacing  $f_{\rm rep} = 1/T$ . This is schematically shown in Figure 2.6, and the frequency of the *n*-th (*n* is an integer) mode is

$$f_n = f_{\rm ceo} + n \times f_{\rm rep}, \tag{2.35}$$

where  $f_{ceo}$  is the carrier-envelope offset frequency, related to the exact

phase and group velocity inside the cavity. Both  $f_{\rm ceo}$  and  $f_{\rm rep}$  are in the rf (10 – 500 MHz) domain. In the optical domain the frequency range of the comb is around  $f_n \approx 10^{15}$  Hz, and we find that typically  $n \approx 10^6$ . In this way the frequency comb relates the optical frequency  $f_n$  to rf frequencies  $f_{\rm ceo}$  and  $f_{\rm rep}$ , which are easily compared to the SI standard. The mode number n can be calculated from other measurements or calibrations, thus the frequency comb allows direct measurement of optical frequencies with over 12-digit absolute accuracy [215, 216], depending on the reference standard (i.e. a rubidium or caesium atomic clock).

The first frequency combs were build using a Ti:sapphire gain medium and therefore operated at the 800 nm wavelength regime, but nowadays they are extending into the XUV regime [29, 217] up to the mid-infrared [218, 219] with appreciable powers to do spectroscopy. Additionally, frequency combs with fiber-based technology are now commercially available in the 1300-1600 nm (and frequency-doubled) wavelength range due to developments in the optical telecommunications industry.

For the experiments discussed here we use an erbium-doped fiber laser frequency comb (Menlo Systems, hosted by the Ultrafast Laser Physics and Precision Metrology group) operating in the 1500 nm range with a bandwidth of 100 nm. The optical range of the frequency comb is extended with an optical amplifier and photonic crystal fiber to allow frequency doubling for a secondary comb operating in the 550-900 nm range. For the measurement on the  $2 \, {}^{3}S_{1} - 2 \, {}^{1}P_{1}$  transition (see Chapter 3) the  $f_{ceo}$  and  $f_{rep}$  of the comb were referenced to a GPS-controlled rubidium clock (FS725 by Stanford Research Systems), which provided a  $10^{-12}$  long term stability. At the time of the line shape measurements on the  $2 \, {}^{3}S - 2 \, {}^{1}S$  transition (see Chapter 4) the rubidium clock was upgraded to a stand alone caesium clock (CsIII Model 4310B by Symmetricom) with a long term stability of  $< 10^{-13}$ . This is required for a sub-kHz absolute accuracy on the determination of the  $2 \, {}^{3}S - 2 \, {}^{1}S$ transition frequency.

### Ultrastable laser

Whereas a frequency comb spans a broad wavelength range, progress has also been made in so-called ultrastable lasers. Ultrastable lasers are generally characterized by a very high quality factor  $\nu_0/\delta\nu$ , where  $\nu_0 \approx 10^{15}$  Hz is the nominal optical oscillation frequency and  $\delta \nu$  the frequency noise<sup>9</sup>. The high stability of these lasers is achieved by locking them to a very stable reference, generally a cavity [221]. As the locked laser frequency depends on the exact cavity length, these cavities are designed to be insensitive to mechanical vibrations [222] and temperature fluctuations [223, 224], and are kept in vacuum to reduce acoustic vibrations. With these efforts stabilities (i.e. the inverse of the quality factor) of  $10^{-15}$  can be achieved at the timescale of 1 s (i.e.  $\delta \nu < 1$  Hz) which is four to five orders of magnitude better than the stability of a rubidium or caesium clock reference signal at the same timescale. An ultrastable laser will always have a long-term drift due to slow drift in the cavity length or chemical processes affecting the cavity length, and should be referenced to a clock to correct for this drift. Combining ultrastable lasers with even better frequency references (i.e. ultranarrow atomic transitions in the optical regime which have extremely high quality factors) is a pathway to a new generation of clocks surpassing the current SI standard by multiple orders of magnitude [225–229]. Alternatively, direct comparison of frequency ratios of different transitions measured with ultrastable lasers allows determination of relative changes of fundamental constants [64, 65, 230, 231].

The Ultrafast Laser Physics and Precision Metrology group also has an ultrastable laser (ORS1500 by Menlo Systems) in a separate temperaturecontrolled room near the optical frequency comb setup. The laser operates at 1542 nm with a linewidth specified to be below 2 Hz, resulting in a short term (1-10 s) stability of around  $10^{-15}$ . The linear drift of the ultrastable laser is specified to be better than 0.15 Hz/s, and we have observed a drift of ~ 0.023 Hz/s over almost a year.

## Transfer lock

Apart from the small bandwidth of the feedback loop used in the previous measurement [31, 190], direct frequency stabilization to the erbiumdoped fiber frequency comb is not optimal as the comb modes themselves have a linewidth of ~ 100 kHz limited by the stability of  $f_{ceo}$ . With the

<sup>&</sup>lt;sup>9</sup>The phrase 'frequency noise' is very ambiguous as it has to referred to a specific time scale and type of noise [220].

new ultrastable laser system available in our laboratory we can circumvent this by implementing a so-called 'transfer lock' scheme.

The phrase 'transfer lock' or 'transfer oscillator' refers to the basic principle of the technique; by creating a phase-locked loop between an ultrastable laser and a less stable laser, the stability of the ultrastable source will be transferred to the latter. An example of this technique is dual locking to a single Fabry-Pérot cavity [232, 233]. Other experiments rely on the reduction of the linewidth of frequency comb modes [234, 235]; locking of a laser to a stable comb mode also constitutes a transfer lock. The latter method is interesting as the large bandwidth of the frequency comb allows for flexible design with respect to the wavelength of the ultrastable laser and the less stable laser [236, 237]. One can also imagine a transfer lock scheme involving an ultrastable laser at an optical frequency and an atomic clock at a microwave frequency through  $f_{ceo}$  and  $f_{rep}$  [238].

An even simpler scheme is to use a frequency comb simply as the reference oscillator which allows comparison of two other oscillators by creating a so-called 'virtual beatnote' between the individual beatnotes of the lasers with the comb. This transfer lock scheme can be designed in such a way that the frequency comb does not add any noise in the process, simply providing a common oscillator to compare two other optical frequencies [239]. This scheme has already shown its usefulness in various high-precision measurements [65, 240, 241].

In our experiment we use a simplified version of the transfer lock scheme as suggested by Telle [239], as we currently do not need to get a spectroscopy laser linewidth of ~ 1 Hz, but < 10 kHz would already suffice, with considerable ease of implementation compared to the full scheme.

### Creating a 'virtual beatnote'

By spatially overlapping the optical modes from two laser sources with different optical frequencies  $f_1$  and  $f_2$ , the total electric field will oscillate with the sum  $(f_1 + f_2)$  and difference  $(f_1 - f_2)$  frequencies and associated higher modes. If  $f_1$  and  $f_2$  are close enough, even if they are in the optical frequency domain, the difference frequency  $f_1 - f_2$ can be in the radio- or microwave-frequency domain and can be measured electronically using a photodiode (as long as  $f_1 - f_2$  is within the bandwidth of the device). Using the proper electronic filters and amplifiers, the lowest measured difference frequency is generally called the 'beatnote'  $f_b = f_1 - f_2$ .

Creating a beatnote between a frequency comb and a single-frequency laser  $f_1$  produces a beatnote  $f_{b,1}$  following the resonance condition

$$f_1 = f_{\text{ceo}} + n_1 \times f_{\text{rep}} \pm f_{b,1},$$
 (2.36)

where  $f_{b,1}$  is the lowest frequency difference between  $f_1$  and the nearest comb mode and can be negative. If a wavemeter with sub- $f_{\rm rep}/2$ frequency accuracy is available, the absolute mode number  $n_1$  can be determined and the absolute frequency of the single-frequency laser is known. Producing a beatnote with a second single-frequency laser provides a second beatnote  $f_{b,2}$  defined by

$$f_2 = f_{\rm ceo} + n_2 \times f_{\rm rep} \pm f_{b,2},$$
 (2.37)

where the absolute mode number  $n_2 \neq n_1^{10}$ . As both  $f_{b,1}$  and  $f_{b,2}$  are in the rf domain, we can mix them again and filter the difference frequency to obtain the virtual beatnote  $f_{\text{virt}}$ :

$$f_{\rm virt} = f_{b,1} - f_{b,2},\tag{2.38}$$

$$f_{\text{virt}} = (f_1 - f_2) - (n_1 - n_2) \times f_{\text{rep}}.$$
 (2.39)

Note that the carrier-offset frequency already dropped out perfectly in  $f_{\rm virt}$ , assuming that the rf signals are still coherent when they are mixed. Furthermore this assumes that  $f_{b,1}$  and  $f_{b,2}$  have the same sign, otherwise the carrier-offset frequency gets doubled in the virtual beatnote and the noise correspondingly doubles instead of being cancelled. The virtual beatnote exists of a detuning term  $f_1 - f_2$  and a residual frequency correction from the repetition rate of the frequency comb, and is equivalent to  $(f_1 - f_2) \operatorname{Mod}(f_{\rm rep})$ . Establishing a phase-locked loop with the virtual beatnote to a stable reference oscillator and providing feedback to the spectroscopy laser, will effectively phase-lock the spectroscopy laser to the ultrastable laser thereby creating the transfer lock.

In the full scheme by Telle [239] the beatnote frequencies are mixed in such a way that the second term on the right hand side of Eq. 2.39

<sup>&</sup>lt;sup>10</sup>If  $n_2 = n_1$  it is easier to directly create a beatnote between the two lasers.

does not add any noise in the virtual beatnote, making it completely independent of the frequency comb. In our case we do not compensate for this, but as  $\Delta n/n \approx 1\%$  for the two wavelengths in our experiment (1542 nm and 1557 nm) the residual noise is acceptable for achieving our spectroscopy laser linewidth goal.

#### Transfer lock setup

An overview of the full transfer lock scheme is shown in Figure 2.7. The fiber-coupled output of the frequency comb is split from the full spectrum at the relevant wavelengths of 1542 nm and 1557 nm using two fiber Bragg gratings specified for a 1 nm bandwidth around these wavelengths. The optical paths between the different lasers are all fiber-coupled and mixed using fused fiber couplers such that they have perfect mode overlap. The beatnotes between the two lasers are separately measured by VU-constructed ac-coupled and fiber-coupled photodiodes with 150 MHz bandwidth. The frequency comb is configured such<sup>11</sup> that the ultrastable laser beatnote  $f_{\rm stab} \approx 60$  MHz and the spectroscopy laser (Koheras Adjustik E15 by NKT Photonics, linewidth specified < 1 kHz<sup>12</sup>) beatnote is  $f_{\rm spec} \approx 100$  MHz.

After bandpass-filtering and amplification, the spectroscopy laser beatnote is down-mixed with a direct digital synthesizer (DDS, AD9912 by Analog Devices) at a frequency of  $f_{\text{DDS}} \approx 30 \text{ MHz}$  to a new spectroscopy laser beatnote of  $\tilde{f}_{\text{spec}} = f_{\text{spec}} - f_{\text{DDS}} \approx 70 \text{ MHz}$ . The addition of the DDS allows for external control of the absolute spectroscopy laser frequency without breaking the phase-locked loop. The down-mixed beatnote is split by a -3 dB splitter to be monitored by a zero-dead time counter (FXM50 by K+K Messtechnik) for absolute frequency calibration of the spectroscopy laser, and the other port is mixed with the ultrastable laser beatnote to obtain the virtual beatnote

<sup>&</sup>lt;sup>11</sup>If the laser frequencies are known, the required repetition rate can be numerically calculated by simultaneously solving beatnote Eqs. 2.36 and 2.37 with chosen frequencies for the virtual beatnote (Eq. 2.39) and individual beatnotes  $f_{\text{stab}}$  and  $f_{\text{spec}}$  as boundary condition.

 $<sup>^{12}{\</sup>rm The}$  short-term linewidth of the NP Photonics laser previously used for spectroscopy is specified to be 5 kHz.



Figure 2.7 – Schematic overview of the transfer lock setup. Using fiber optics, the optical frequencies of the three lasers are mixed on two separate ac-coupled photodiodes. The beatnotes are band-pass filtered and amplified; the nominal frequencies are shown in the Figure. The spectroscopy laser beatnote is separately down-mixed with a direct digital synthesizer ('DDS') to allow for external control of the absolute laser frequency in the phase-locked loop. This beatnote (at  $\sim 70 \text{ MHz}$ ) is split by a - 3 dB splitter to be measured by a zero-dead time counter ('Counter') and mixed with the ultrastable laser beatnote to obtain the virtual beatnote at  $\sim 10$  MHz. The virtual beatnote is discretized using a digital clock divider ('DCD') simultaneous with the 10 MHz reference signal from the atomic clock. Both discretized signals are fed in a phase-frequency detector ('PFD') which produces an error signal based on the relative phases. A home-built low-noise analog proportional-integral ('PI') controller provides feedback on the piezoelectric controller of the spectroscopy laser, thereby achieving a phase-locked loop. The symbols for the different rf components (all from Minicircuits) are explained below the figure.
$f_{\rm virt} = \tilde{f}_{\rm spec} - f_{\rm stab} \approx 10$  MHz. The virtual beatnote is monitored by an identical zero-dead time counter and sent to a digital clock divider. The VU-constructed digital clock divider discretizes the analog oscillating signal to a block-pulse signal (with optional fractional time-division) for better comparison in the phase-frequency detector. Simultaneously the 10 MHz reference signal from the atomic clock is sent in the digital clock divider and consequently the phase-frequency detector, of which the basic principle is explained in Ref. [242]. The VU-constructed phasefrequency detector is able to follow up to  $32\pi$  cycle slips, but nominally operates in a  $\pm \pi$  phase range for which it outputs 0 - 4V (i.e. the phases are 'locked' at an output voltage of 2 V). The error signal from the phase-frequency detector is fed in a low-noise analog proportionalintegral controller with a bandwidth of  $\sim 25 \,\mathrm{kHz}$ , which is sufficient to cover the 20 kHz bandwidth of the piezoelectric controller of the spectroscopy laser. With a frequency sensitivity of the laser with respect to the piezoelectic controller voltage of 16 kHz/mV, the electronic noise of the feedback signal should be  $< 100 \ \mu V$  which is achieved by the custom built PI controller<sup>13</sup>.

#### Frequency metrology infrastructure

A full overview of the metrology infrastructure in the laboratory is discussed to complete the description of the new spectroscopy laser and frequency stabilization scheme, as shown in Figure 2.8. The spectroscopy laser is situated near the frequency comb to minimize the length of the feedback loop in the transfer lock, and the spectroscopy light is transported to the He<sup>\*</sup> experiment using a ~ 80 m uncompensated fiber link. The ultrastable laser and caesium clock are located in a small temperature-stabilized and quiet room (the 'isolated room') inside the Ultrafast Laser Physics and Precision Metrology laboratory, and another uncompensated fiber link of ~ 25 m allows for a beatnote at the frequency comb.

The external control of the absolute frequency of the spectroscopy laser, set by the in-loop DDS, is controlled by a local computer near the frequency comb. This computer also communicates with the frequency

 $<sup>^{13}\</sup>mathrm{We}$  thank Tim Kortekaas of the electronics workshop who designed and built the controller.



**Figure 2.8** – Schematic overview of the full metrology infrastructure, including the spatial separation between the different laboratories. Within the Ultrafast Laser Physics and Precision Metrology group laboratory, both the ultrastable laser and caesium clock are placed in a temperature-stabilized and quiet 'isolation room'.  $A \sim 25$  m uncompensated fiber link connects the ultrastable light to the frequency comb in the main room. The spectroscopy laser is situated near the frequency comb and transfer-locked to the ultrastable laser ('TL'), which is shown in detail in Figure 2.7.  $A \sim 80$  m uncompensated fiber link between the labs transports the spectroscopy light to the experiment. A secondary computer ('PC comm.') interfaces between the labs and allows readout of all relevant frequencies, and controls the DDS within the TL to set the absolute frequency of the spectroscopy laser. All frequency measurement devices or sources are referenced to the caesium clock.

comb and the counters; all relevant frequencies and settings can therefore be documented during the measurements. Communication between the two laboratories is mediated over the local network of the university to a computer at the He<sup>\*</sup> experiment. This computer periodically communicates with the computer controlling the full experiment, but the datastream is small enough to not interfere with the experiment.

All frequency counters and sources relevant for the high-precision spectroscopy (in both laboratories) are referenced to the caesium clock. As the GPS-disciplined rubidium clock is still available, a separate computer logs the pulse-per-second signal of both clocks to enable traceability of the caesium clock (not shown in Figure 2.8). The long-term stability of the rubidium clock is only  $1 \times 10^{-12}$ , and therefore only allows us to observe a gross instability of the caesium clock. The caesium clock stability floor is specified to be  $< 5 \times 10^{-14}$ , which is more than sufficient for the future accuracy goals of the He<sup>\*</sup> spectroscopy<sup>14</sup>.

#### Transfer lock performance

Simultaneous monitoring of two beatnotes by zero-dead time frequency counters is necessary to determine the absolute frequency of the spectroscopy laser during the measurements, and the difference between two beatnotes is used to observe cycle-slips in the phase-locked loop. Essentially any two of the three  $(f_{\text{stab}}, f_{\text{spec}}, \text{ or } f_{\text{virt}})$  frequencies need to be monitored to make sure that cycle-slips can be observed. E.g. we could have chosen to monitor  $f_{\text{stab}}$  in stead of  $f_{\text{virt}}$ , as shown in Figure 2.7, but the current setup is more convenient as we also observe  $f_{\text{virt}}$  on a spectrum analyzer to quickly see if a phase-locked loop is established.

An example of an almost 17-hour continuous measurement of the in-loop virtual beatnote (in this specific measurement obtained by subtracting  $f_{\text{stab}}$  and  $\tilde{f}_{\text{spec}}$ ) is shown<sup>15</sup> in Figure 2.9, showing that the optical frequencies of the spectroscopy laser and ultrastable laser are phase-locked

<sup>&</sup>lt;sup>14</sup>There currently is an ultrastable optical link from the Dutch metrology institute VSL to the Dutch National Institute for Subatomic Physics Nikhef in Amsterdam [243]. Extension of this link to the LaserLaB would allow unambiguous traceability of the caesium clock beyond the  $1 \times 10^{-14}$  level.

<sup>&</sup>lt;sup>15</sup>We display the measured frequencies in a density plot as the datapoints are too close to visualize individually. The raw measurement data itself of course exists of discrete datapoints taken at a 1 s interval.

to within a fraction of 1 Hz at the integration time scale of one second. Any cycle-slips would immediately occur as a gross frequency deviation from this distribution, and are therefore easy to detect in the postprocessing of this data.

For timescales shorter than one second we look at the rf spectrum of the virtual beatnote centered around the reference frequency of 10 MHz and construct the power spectral density (PSD) of the frequency components, as shown in Figure 2.10. Indeed we see the  $\sim 20 \,\mathrm{kHz}$  servo bandwidth of the feedback loop due to the bandwidth of the piezoelectric controller of the spectroscopy laser. Near the carrier frequency we observe a regular sideband structure as harmonics of 50 Hz, indicating possible amplitude modulation caused by the voltage supplies. This is currently no limiting factor for our experiments as the linewidth of our laser will be in the kHz regime, and most sidebands are more than 20 dBc below the carrier frequency and can be safely ignored. A PSD of the spectroscopy laser beatnote is not shown as it is fully dominated by noise from the frequency comb, and an accurate beatnote of the free running spectroscopy laser was impossible to acquire as it has a specified short term linewidth of  $< 1 \, \text{kHz}$ , but the frequency drifts at  $\sim 1 \, \text{MHz/s}$ . Figure 2.11 shows the absolute beatnote of the spectroscopy laser and the frequency comb while the spectroscopy laser is transfer-locked to the ultrastable laser, and belongs to the same dataset as shown in Figure 2.9. The beatnote has a slow linear drift as it is locked to the ultrastable laser and the extracted linear drift is  $\sim 0.03 \, \text{Hz/s}$ , similar to the average drift on a yearly basis. Subtracting this linear drift we observe again a Gaussian distribution of the beatnote but now with a  $\sim 7 \, \text{kHz}$  FWHM. This is four orders of magnitude larger than the width of the virtual beatnote and caused by the linewidth of the frequency comb mode, underlining the advantage of the transfer lock to the ultrastable laser in stead of frequency stabilizing to the frequency comb. Figure 2.11 also reveals a periodic modulation on the beatnote with a period of  $\sim 3$  hours (confirmed by a Fourier transform of the data set) which is not visible in the virtual beatnote. This is caused by a slow feedback loop in the rubidium clock leading to a slow modulation of  $f_{\rm rep}$  of the frequency comb. The caesium clock does not have such a feedback loop, as can be see in Figure 2.12. It additionally has a better short-term stability than the rubidium clock.



Figure 2.9 – In-loop virtual beatnote frequency  $f_{virt}$  measured by subtracting the separately measured beatnote between the frequency comb and the spectroscopy laser and ultrastable laser, respectively. The virtual beatnote is phase-locked to the 10 MHz reference signal from the caesium clock, which is therefore subtracted from the frequency. (Left) continuous measurement of the virtual beatnote over  $6 \times 10^4$  s without a single cycle slip, and a histogram of the same data set (right) shows a Gaussian distribution with  $\sigma = 0.0907(3)$  Hz (FWHM = 0.2136(7) Hz).



**Figure 2.10** – In-loop power spectral density (PSD) of the virtual beatnote shown for a frequency span of 3 kHz (left) and 40 kHz (right). Both spectra are averaged over five scans and acquired with a 1 Hz bandwidth. The apparent servo bandwidth is  $\sim 20 \text{ kHz}$  as expected from the piezoelectric controller bandwidth. Higher harmonics of 50 Hz electronic noise can be observed and are all below -20 dBc.



Figure 2.11 – Beatnote between the spectroscopy laser and the rubidium clock-referenced frequency comb while the spectroscopy laser is transfer-locked to the ultrastable laser (i.e. complementary to the dataset as Figure 2.9). (Left) linear drift-corrected  $(-0.0296(7) \text{ Hz/s}, \langle f_b \rangle = 68\,039\,788(12) \text{ Hz})$  beatnote, and a histogram of the same data set (right) shows a Gaussian distribution with  $\sigma = 2.895(8) \text{ kHz}$  (FWHM = 6.82(2) kHz).



Figure 2.12 – Beatnote between the spectroscopy laser and the caesium clock-referenced frequency comb while the spectroscopy laser is transfer-locked to the ultrastable laser. (Left) linear drift-corrected  $(-0.0099(6) \text{ Hz/s}, \langle f_b \rangle = 75942824(11) \text{ Hz})$  beatnote, and a histogram of the same data set (right) shows a Gaussian distribution with  $\sigma = 2.089(7) \text{ kHz}$  (FWHM = 4.92(2) kHz). The frequency axes are identical to Figure 2.11 showing that the beatnote has better short-term stability and no oscillations as compared to the rubidium clock-referenced frequency comb.



Figure 2.13 – Overlapping Allan deviation calculated from the driftcorrected beatnote as shown in Figure 2.12. After an averaging time of 10 s the Allan deviation decreases as  $\tau^{\alpha}$ , with  $\alpha = -0.49(1)$  (dashed line). The gray triangles for  $\tau > 10^3$  s show the long-term instability of the beatnote without drift-correction.

The long term stability of the beatnote can also be evaluated by calculating the overlapping Allan deviation [220], which is a measure of the fractional stability for various averaging times  $\tau$ . For the same dataset as shown in Figure 2.12 the corresponding overlapping Allan deviation is shown in Figure 2.13. The figure shows that the initial fractional stability of the spectroscopy laser frequency is  $\sim 7 \times 10^{-12}$  (i.e.  $\sim 1.3$  kHz) and after 10 s of averaging time starts to decrease as  $\tau^{-1/2}$ , which means that the signal is dominated by white frequency modulation noise [220]. We see that we can average down to a stability of  $2 \times 10^{-13}$  in  $10^4$  s (i.e. under three hours), which is sufficient to obtain a sub-100 Hz absolute accuracy on the spectroscopy laser frequency.

#### Spectroscopy laser linewidth

The goal of implementing the transfer lock is to reduce the former spectroscopy laser linewidth of  $\sim 100 \text{ kHz}$  to < 10 kHz. Based on the imple-

mented transfer lock and the observation that the width of the beatnote of the spectroscopy laser with the frequency comb has decreased, we can assume that the linewidth has been significantly reduced. In this section several contributions to the final linewidth of the spectroscopy laser at the experiment are listed, and how they can be improved in the future. There are several contributions to the linewidth of the spectroscopy laser. As shown in Eq. 2.39, the virtual beatnote as used in our experiment still carries a residual contribution of the comb modes as the mode numbers of the two beatnotes are not equal. The relative difference of the two modenumbers  $\Delta n/n \approx \Delta \lambda/\lambda \approx 1\%$ , and with a comb mode linewidth of ~ 100 kHz, a ~ 1 kHz linewidth broadening effect is expected as the full modenumber-compensated Telle scheme [239] is currently not used.

Furthermore the fiber links in the laboratories are not yet stabilized. For the fiber link of the ultrastable laser to the frequency comb ( $\sim 25 \,\mathrm{m}$ ) linewidth broadening to 100 Hz of the ultrastable laser light is estimated. The spectroscopy laser will adopt this linewidth in the transfer lock scheme. In the near future this fiber link will be actively stabilized, but it is not yet a limiting factor of our spectroscopy laser linewidth.

Another contributing factor to the linewidth of the spectroscopy laser is the uncompensated fiber link from the frequency comb laboratory to the experiment ( $\sim 80 \,\mathrm{m}$ ). Delayed self-heterodyne beatnote measurements of the roundtrip between the two labs show a  $\sim 4 \,\mathrm{kHz}$  FWHM broadening, meaning that a single trip will broaden the linewidth by  $\sim 2 \,\mathrm{kHz}$  at the 1 s timescale.

Finally we also have electronic noise from the feedback system. Although the PI controller is designed to be low-noise (<  $100 \mu$ V), the linewidth can still broaden due to this noise. This can be observed in a delayed self-heterodyne beatnote measurement using a 100 km fiber link available in the laboratory. Due to its large length, the fiber link is very sensitive to acoustic noise, and the pure linewidth of the laser could not be determined. However, the observed acoustic-noise broadened linewidth can be compared in various circumstances (e.g. for a free running laser, 'tight' PI settings, and 'loose' PI settings) to determine additional broadening effects. These measurements show that the increase of the laser linewidth due to the electronics is ~ 1 - 2 kHz at the 1 s timescale, and would increase (but not oscillate) for higher PI settings (i.e. in a 'tight' lock configuration).

All contributions have their own spectral behavior and would require full spectral analysis to obtain the effective linewidth of the spectroscopy laser [244]. To obtain an estimate the contributions are simply added and give an estimated 4 - 5 kHz linewidth of the spectroscopy laser for the experiment. The spectroscopy laser linewidth can be extracted from the line shape measurements discussed in Chapter 4, and we find a linewidth of 4.5(3) kHz, which is in agreement with the estimation.

From the inventory of linewidth broadening sources one can conclude that stabilization of the fiber link would be the next step to improve the spectroscopy laser linewidth by a factor of two. Reduction of the frequency comb mode linewidth by stabilizing the frequency comb to the ultrastable laser [234, 236] or by implementing the full modenumbercompensated Telle transfer locking scheme [239] will then be the path towards a sub-kHz linewidth.

# CHAPTER 3

# High-precision spectroscopy of the $2 \ {}^{3}S_{1} \rightarrow 2 \ {}^{1}P_{1}$ transition

#### Abstract

We have measured the forbidden 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition at 887 nm in a quantum degenerate gas of metastable <sup>4</sup>He atoms confined in an optical dipole trap. The determined transition frequency is 338 133 594.4 (0.5) MHz, from which we obtain an ionization energy of the 2  ${}^{1}P_{1}$  state of 814 709 148.6 (0.5) MHz. This ionization energy is in disagreement by  $> 3\sigma$  with the most accurate quantum electrodynamics (QED) calculations available. Our measurements also provide a determination of the lifetime of the 2  ${}^{1}P_{1}$  state of 0.551 (0.004)<sub>stat</sub> ( ${}^{+0.013}_{-0.000}$ )<sub>syst</sub> ns, which is the most accurate determination to date and in excellent agreement with theory.

# **3.1** Introduction

Quantum electrodynamics is one of the most thoroughly tested theories in physics. From QED theory and accurate measurements, the fine structure constant [20, 21], the Rydberg constant [92], nuclear charge radii [31, 95] and the electron mass can be deduced [83]. It can also provide accurate ionization energies for one- and two-electron atoms. To

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test QED, both highly accurate calculations and high-precision experimental data are required. Few-body systems such as the hydrogen atom and helium atom are candidates that fulfill both criteria. Testing and applying QED in these systems has led to surprising results in recent years. An example is the  $7\sigma$  discrepancy in the proton size derived from muonic hydrogen Lamb shift measurements and the accepted CODATA value, also referred to as the proton radius puzzle [95, 97]. Recent measurements of the helium  $2 {}^{3}S \rightarrow 2 {}^{1}S$  transition at 1557 nm [31] and the 2  ${}^{3}S \rightarrow 2 {}^{3}P$  transitions at 1083 nm [44, 105] disagree by  $4\sigma$  in the determination of the helium isotopic nuclear size difference, even with a recent recalculation of QED effects in the isotope shift [27]. These measurements provide a unique comparison with nuclear size measurements in the muonic helium ion, developed to help solve the proton radius puzzle [100]. Also recent experiments with trapped highly charged heliumlike ions show a significant discrepancy with QED theory,  $3\sigma$  for  $Ti^{20+}$  and growing as  $Z^3$  [245]. The validity of these results has been lively discussed [246, 247], and other experiments contradict the results [248].

In particular for the low-lying states with low angular momentum, accurate measurements of the ionization energies (IE) in helium have allowed stringent tests of two-electron QED [29, 31, 32, 36, 38, 39, 44]. A schematic overview of the lowest states of helium together with transition wavelengths mentioned in this Chapter are shown in Figure 3.1. In comparing the experimentally determined IE to QED calculations, a discrepancy of 6.5 (3.0) MHz in the 2  ${}^{1}P_{1}$  IE was identified by Drake and Pachucki [26, 55, 57]. This discrepancy is based on a measurement of the 2  ${}^{1}P_{1} \rightarrow 3 {}^{1}D_{2}$  transition frequency with 3 MHz accuracy by Sansonetti and Martin in 1984 [249]. As the QED calculation of this IE is accurate to 0.4 MHz [26], a more accurate measurement should be able to determine whether this discrepancy still stands. Recently, two new determinations of the 2  ${}^{1}P_{1}$  IE were reported by Luo *et al.* based on the measurements of the 2  ${}^{1}S_{0} \rightarrow 2 {}^{1}P_{1}$  [33] and 2  ${}^{1}P_{1} \rightarrow 3 {}^{1}D_{2}$  [34] transition frequencies. As these transitions are electric dipole-allowed, the measurements could be done using saturated absorption spectroscopy in an rf discharge cell. The extracted ionization energies for the 2  ${}^{1}P_{1}$ state disagree with QED theory at the  $3.5\sigma$  level.

In this work we report the direct measurement of the forbidden 2  ${}^{3}S_{1} \rightarrow$ 



**Figure 3.1** – Schematic overview of the lowest levels in <sup>4</sup>He and the transitions that are mentioned in this chapter. The 2  ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{2}$  transition at 1083 nm is used for laser cooling and trapping.

2  ${}^{1}P_{1}$  transition at 887 nm in a quantum degenerate gas (QDG) of metastable 2  ${}^{3}S_{1}$  state helium (denoted He<sup>\*</sup>, lifetime  $\approx$  7800 s) atoms confined in an optical dipole trap (ODT). The advantage of performing spectroscopy in an ODT is the ability to probe very weak transitions and the simultaneous reduction and characterization of systematic effects to the kHz level [31]. As the theoretical natural linewidth of this transition is 287 MHz [250], the accuracy of our measurement is limited by statistics rather than by systematic effects. Combined with the accurately known IE of the 2  ${}^{3}S_{1}$  state this measurement of the transition frequency enables a determination of the 2  ${}^{1}P_{1}$  IE. The measured line shape of the transition allows for an accurate determination of the lifetime of the 2  ${}^{1}P_{1}$  state. This method does not require the branching ratios of decay channels, which is the main problem in fluorescence measurements of the lifetime [251–255], and the only dominant broadening effect in our experiment can be calculated using the optical Bloch equations.

# 3.2 Experimental setup

The 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition is forbidden as it violates conservation of spin. Due to a small mixing of the  $2 {}^{1}P_{1}$  and  $2 {}^{3}P_{1}$  states, the effective eigenstate is  $\sqrt{1-\epsilon^2} |2^{-1}P_1\rangle + \epsilon |2^{-3}P_1\rangle$  with  $\epsilon \approx 3 \times 10^{-4}$  [256]. It is this slight mixing with the triplet states which enables the electric dipole transition with an Einstein A coefficient of  $1.4423 \text{ s}^{-1}$ , which is seven orders of magnitude weaker than regular dipole-allowed transitions in the helium atom [56]. Therefore this transition has, to our knowledge, never been observed before. In order to obtain a good signal with reasonable laser power, the atoms need to be probed on a timescale of about 1 s, and we achieve this by trapping a QDG of  ${}^{4}\text{He}^{*}$  atoms in an ODT. For this we use the same experimental setup as used to measure the doubly forbidden  $2 {}^{3}S \rightarrow 2 {}^{1}S$  transition [31]. We produce a QDG consisting of a thermal gas and a Bose-Einstein condensate (BEC) in a crossed-beam ODT, which is created using an NP Photonics Scorpio fiber laser operating at a wavelength of 1557.3 nm. Details on the production and physics of ultracold metastable gases can be found in [132] and Chapter 2. The advantage of using a QDG for spectroscopy is the small ODT trap depth required to trap the gas, which minimizes systematic transition frequency shifts. The ODT is kept shallow at a depth of about 1.3  $\mu$ K, and after thermalization the temperature of the gas is approximately 0.2  $\mu$ K. We apply a small homogeneous magnetic field in the ODT to maintain spin polarization of the gas, which is required to have a trap lifetime > 10 s. The small Zeeman shift is directly measured using rf transitions between the 2  ${}^{3}S_{1}$   $m_{I} = +1, 0, -1$  states with kHz accuracy and therefore does not provide a limitation for our experimental accuracy [31].

A schematic overview of the ODT and the metrology infrastructure is



**Figure 3.2** – Schematic overview of the setup. The crossed dipole trap is created using a fiber laser and the trapping beam power is measured using a power meter. The spectroscopy light is generated using a Ti:sapphire ring laser. The beatnote between the spectroscopy laser and the frequency comb is measured with an avalanche photodiode (APD) connected to a frequency counter and digitally sent to the lab computer. The computer then calculates and sends a proportional-integral (PI) feedback signal to the Ti:sapphire laser to stabilize the spectroscopy laser frequency. The lab computer also interfaces with the frequency comb, enabling us to register and control the frequency comb settings.

shown in Figure 3.2. The probe beam is generated using a Coherent 899-21 Ti:sapphire laser with an output power of 0.4 W at 887 nm. During the measurements the wavelength is registered using an ATOS wavemeter. Simultaneously we use an erbium-doped fiber laser frequency comb that is stabilized using a GPS-controlled rubidium clock to create a beatnote with the probe laser [31]. Combining the wavemeter data with the beatnote data provides the absolute frequency of the probe laser.

Additionally, we stabilize the Ti:sapphire laser frequency to the frequency comb using a proportional-integral (PI) control loop. We control the Ti:sapphire laser frequency by keeping the beatnote frequency constant and scanning the repetition rate of the frequency comb. Due to the relatively slow loop time of 30 ms of the PI control loop, our laser has a Gaussian line shape with an average FWHM of approximately 1 MHz, with an accuracy of < 5 kHz, during the measurements.

In our experiment we measure a line scan over the resonance using 90 individual measurements with a frequency interval of 20 MHz. For every individual measurement, a new QDG sample has to be produced. Once the QDG is loaded in the ODT, an approximately 50 mW probe beam excites the atoms to the  $2 {}^{1}P_{1}$  state during 1 s. The excited atoms decay in 0.55 ns to the 1  ${}^{1}S_{0}$  state and leave the trap. Then the ODT is turned off and the remaining atoms fall due to gravity and hit a microchannelplate (MCP) detector. The MCP current is measured to determine the time-of-flight (TOF) distribution of the atoms. This TOF distribution is fit using a bimodal distribution, which describes the momentum distribution of the BEC and thermal fractions. From the fit we obtain the atom number of both fractions, the temperature of the thermal fraction and the chemical potential of the BEC. Having measured the remaining total number of atoms at all 90 points, the line shape can be normalized by the far off-resonance remaining atom number. In this way we reconstruct the absorption line shape of transition.

# 3.3 Transition line shape

As the lifetime of the excited state is only 0.55 ns and the measured transition is too weak for stimulated emission to occur, the excited atoms decay to the 1  ${}^{1}S_{0}$  ground state before they even have the chance to

move out of the trap potential (which is anti-trapping for the 2  ${}^1P_1$ state). The recoil kick of the emitted 58-nm photon is large enough for the ground state atom to leave the trap. In this process we ignore the alternative decay path 2  ${}^{1}P_{1} \rightarrow 2 {}^{1}S_{0} \rightarrow 1 {}^{1}S_{0}$  which also takes place but at a branching ratio of 0.1% compared to the single-photon decay. Still, as the 2  ${}^{1}S_{0}$  state experiences an anti-trapping potential in the optical dipole trap atoms decaying via this channel will be observed as a loss indiscernable from the primary channel. Therefore any atom that is excited, will leave the trap. This means that even if the probe beam is off-resonant all atoms will eventually be lost from the trap. In the limiting case of infinitely long interaction time, one would then observe an infinitely broad transition. In practice the one-body lifetime of the gas is finite ( $\tau \ge 10$  s), but this is not relevant for this experiment as the interaction time is much shorter ( $\sim 1 \, s$ ). This effect leads to broadening which should be corrected for. Therefore we calculate the population of the three states that are involved in this problem and evaluate the population of the initial state as a function of interaction time to see what happens to the linewidth as measured in the experiment.

We use a simple three-level system consisting of the metastable 2  ${}^{3}S_{1}$  state, the excited 2  ${}^{1}P_{1}$  state and the 1  ${}^{1}S_{0}$  ground state. The offresonant scattering rates of the most probable transitions from the 2  ${}^{3}S_{1}$ and the 2  ${}^{1}P_{1}$  states to higher states either due to the 886.6 nm or 1557.3 nm light is negligible (> 10<sup>7</sup> linewidths detuned) compared to the Rabi frequency of the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition. Photo-ionization of the 2  ${}^{3}S_{1}$ state due to reabsorption of a 58-nm photon emitted during the decay of the 2  ${}^{1}P_{1}$  state is also fully negligible [257]. A schematic overview of the three-level system is shown in Figure 3.3, where we have the Einstein A coefficient  $A_{21} = 1.442 \text{ s}^{-1}$  and added two decay channels  $A_{13} = 1.272 \times 10^{-4} \text{ s}^{-1}$ , and  $A_{23} = 1.801 \times 10^{9} \text{ s}^{-1}$  [56]. For linear polarized light, the Rabi frequency in this system is defined as

$$\Omega_R^2 = \frac{3\pi c^2}{\hbar\omega_0^3} A_{21} I_0, \qquad (3.1)$$

where  $I_0 = 2P/\pi w_0^2$  is the intensity of the spectroscopy beam, P the spectroscopy beam power and  $w_0$  the beam waist. The transition frequency is given by  $\omega_0/2\pi$ . For a typical probe beam power of  $P \approx 50 \text{ mW}$  and a beam waist of  $w_0 \approx 2.5 \text{ mm}$ , the Rabi frequency  $\Omega_R \approx 2\pi \times 30 \text{ kHz}$ .



**Figure 3.3** – Schematic of the level scheme as used in the OBE model. The populations of the three atomic states are denoted  $\rho_{11}, \rho_{22}$  and  $\rho_{33}$ , respectively. The different states are coupled through the decay channels  $A_{23}$  and  $A_{13}$  and the spontaneous emission rate  $A_{21}$ . The Rabi frequency  $\Omega_R$  represents the interaction with the probe beam, which can have a detuning  $\Delta$  with respect to the resonance frequency.

The optical Bloch equations (OBEs) describing the populations of the states of this three-level system are based on the OBEs described by Van Leeuwen and Vassen for a similar system in helium [106] and are

$$\dot{\rho}_{11} = A_{21}\rho_{22} - A_{13}\rho_{11} + i\frac{\Omega_R}{2}(\rho_{21} - \rho_{12}),$$
  

$$\dot{\rho}_{22} = -(A_{23} + A_{21})\rho_{22} - i\frac{\Omega_R}{2}(\rho_{21} - \rho_{12}),$$
  

$$\dot{\rho}_{33} = A_{23}\rho_{22} + A_{13}\rho_{11}.$$
(3.2)

We can simplify this system of differential equations by writing  $A_{13} = \Gamma_1$ and  $A_{23} = \Gamma_2$  and applying the approximation  $\Gamma_2 \gg A_{21} \gg \Gamma_1$ . Note that the linewidth of the measured transition  $\Gamma = A_{21} + A_{23} \approx \Gamma_2$  (which is accurate up to 9 digits). The OBEs describing the coherences between the states are

$$\dot{\rho}_{12} = -\left(\frac{\Gamma_1 + \Gamma_2}{2} + i\Delta\right)\rho_{12} + i\frac{\Omega_R}{2}(\rho_{22} - \rho_{11}),$$
  
$$\dot{\rho}_{13} = -\frac{\Gamma_1}{2}\rho_{13},$$
  
$$\dot{\rho}_{23} = -\frac{\Gamma_2}{2}\rho_{23},$$
  
(3.3)

where  $\rho_{ij} = \rho_{ji}^{\dagger}$ . Introducing the parameters  $p = (\rho_{12} + \rho_{21})/2$  and  $q = i(\rho_{21} - \rho_{12})/2$ , we get

$$\begin{split} \dot{\rho}_{11} &= \Omega_R q, \\ \dot{\rho}_{22} &= -\Gamma_2 \rho_{22} - \Omega_R q, \\ \dot{\rho}_{33} &= \Gamma_2 \rho_{22}, \\ \dot{p} &= -\frac{\Gamma_2}{2} p + \Delta q, \\ \dot{q} &= -\frac{\Gamma_2}{2} q - \Delta p + \frac{\Omega_R}{2} (\rho_{22} - \rho_{11}), \\ \dot{\rho}_{13} &= 0. \end{split}$$
(3.4)

The full solution of the population of the initial state,  $\rho_{11}$ , is

$$e^{t\frac{\Gamma_2}{2}}\rho_{11}(t) = \frac{\sqrt{S} + \Gamma_2^2 + 4\Delta^2}{2\sqrt{2}} \cosh\left(\frac{t}{\tau_1}\right)$$
$$+ \frac{\Gamma_2}{\sqrt{2}} \frac{\sqrt{S} + D^+}{\sqrt{S}\sqrt{\sqrt{S} + D^-}} \sinh\left(\frac{t}{\tau_1}\right)$$
$$+ \frac{\sqrt{S} - \Gamma_2^2 - 4\Delta^2}{2\sqrt{2}} \cos\left(\frac{t}{\tau_2}\right)$$
$$+ \frac{\Gamma_2}{\sqrt{2}} \frac{\sqrt{S} - D^+}{\sqrt{S}\sqrt{\sqrt{S} - D^-}} \sin\left(\frac{t}{\tau_2}\right), \qquad (3.5)$$

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where the parameters  $S, D^{\pm}, \tau_1$  and  $\tau_2$  are defined as

$$S \equiv (4\Delta^2 + (\Gamma_2 - 2\Omega_R)^2)(4\Delta^2 + (\Gamma_2 + 2\Omega_R)^2),$$
  

$$D^{\pm} \equiv \Gamma_2^2 - 4\Omega_R^2 \pm 4\Delta^2,$$
  

$$\tau_1 \equiv \frac{2\sqrt{2}}{\sqrt{\sqrt{S} + D^-}},$$
  

$$\tau_2 \equiv \frac{2\sqrt{2}}{\sqrt{\sqrt{S} - D^-}}.$$
(3.6)

We recognize the trigonometric terms that represent the coherent excitation, but in our parameter regime these do not play a significant role as  $\Omega_R \ll \Gamma_2$ . The hyperbolic terms are related to the decoherence caused by the decay channel and represent the loss of atoms from the ODT. Figure 3.4 shows the line shape for our experimental parameters at three different interaction times. It is clear that the line becomes broader and deeper as the interaction time increases, which is caused by the loss of atoms from the trap to the 1  ${}^1S_0$  state. As the function is symmetric in  $\Delta$ , no systematic error is expected in the determination of the absolute transition frequency by using a different (but symmetric) line shape function.

The function  $\rho_{11}(\Delta)$  as shown in Figure 3.4 is a function dependent on many parameters and in our experiment we use a simple Lorentzian line shape function defined as

$$y(x) = C_0 - C_1 \left(\frac{C_2}{2}\right)^2 \frac{1}{(x - C_3)^2 + (C_2/2)^2},$$
(3.7)

where  $C_0$  represents the background,  $C_1$  the amplitude of the Lorentzian (which we also call the 'depletion' and is related to  $\rho_{11}$  as  $C_1 = 1 - \rho_{11}(\Delta = 0)$ ),  $C_2$  is the FWHM and  $C_3$  the center frequency. As  $\rho_{11}(\Delta)$ and a Lorentzian function are both symmetric functions, this has no effect on the determination of the transition frequency. To extract the linewidth from these fits we compare the analytical result  $\rho_{11}(\Delta)$  with a Lorentzian line shape and correct for any deviations between these two functions. The lifetime of the 2  ${}^1P_1$  state leads to a Lorentzian distribution in the frequency domain. The fact that we have a slightly different line shape in our experiment is caused by the way the measurement is



**Figure 3.4** – Population of the initial 2  ${}^{3}S_{1}$  state ( $\rho_{11}$ ) as function of the detuning  $\Delta$  for different probe times t. As we increase the probe time the line shape becomes broader and deeper. This is the broadening effect due to the depletion of atoms in the trap.

performed. However, by directly correcting the fitted Lorentzian distribution for deviations from the analytical line shape, the lifetime of the  $2 {}^{1}P_{1}$  state can still be extracted from these measurements. An example of a Lorentzian fit to the analytical result for typical experimental conditions is shown in Figure 3.5. From the structure in the residuals, we conclude that the Lorentzian is accurate beyond the 1% level, after which it deviates from the analytical model most at the center and around  $\Delta = \pm \Gamma/2$ . This problem is parametrized by calculating the difference between the Lorentzian FWHM and the analytical model FWHM as a function of the on-resonance depletion of the trap (simply called 'depletion'). As the depletion gets larger, the discrepancy between the Lorentzian function and the analytical model increases. For a depletion in the range of 0.4-0.6 (i.e. 40%-60% on-resonance loss of atoms, which corresponds to a  $\rho_{11}(\Delta = 0)$  of 0.6-0.4), the correction factor on the FWHM is on average 0.6% and relevant at our level of accuracy. The correction on the depletion is an order of magnitude smaller and



**Figure 3.5** – Lorentzian fit (white dashed line) to the analytical model and the residuals (black dots). From the residuals, which are at  $10^{-3}$ , we infer that the Lorentzian fit is good to the percent level, but is 'quenched' compared to the analytical model. This means that the magnitude of the central peak is larger in the fit than it actually is, and the linewidth of the Lorentzian fit is slightly smaller than the actual linewidth of the curve.



**Figure 3.6** – Relative increase of the Lorentzian linewidth compared to the natural linewidth  $(\Gamma'/\Gamma)$  as function of the on-resonance depletion of the trap. The points are calculated from Lorentzian fits and the curve is an spline-based interpolation function.

negligible.

The previous two corrections are related to the difference between the Lorentzian line shape and the analytical model. The next issue, and the reason why we need to calculate the OBEs, is the linewidth broadening due to depletion of the trap. By comparing the linewidth  $\Gamma'$  determined by a Lorentzian fit to the 'real' linewidth  $\Gamma$  used in the analytical model, the relative increase in linewidth as function of depletion of the trap is calculated and shown in Figure 3.6. Although the increase in linewidth can be calculated to arbitrary precision, the deviation between the Lorentzian line shape and the analytical model becomes larger than our accuracies for a depletion > 0.75 and the comparison of both models becomes less reliable. However, in our experiments our largest depletion is ~ 0.6 and this effect is still smaller than our corrections. As we determine the trap depletion experimentally, we can calculate the correction for the linewidth as measured in the experiment and obtain the natural linewidth. Any experimental uncertainties in the depletion and the

linewidth are propagated through all aforementioned corrections. The correction is not insignificant, as the linewidth is increased on average by 23% just because of depletion of the trap.

Finally, asymmetric broadening effects due to the finite size of the QDG in the ODT and due to the momentum distribution of the gas [203, 258] are below 50 kHz<sup>1</sup> and therefore negligible both compared to the linewidth of the transition and the statistical accuracy of the frequency determination.

# 3.4 Systematic effects

Due to the large (287 MHz) linewidth of the transition, the accuracy in the transition frequency determination is limited by statistics rather than systematics. As will be shown in Section 3.5, the statistical uncertainty in the determined transition frequency is 0.5 MHz. The systematic effects are known which much better precision and therefore do not contribute to the final error budget, but will be discussed here.

#### Recoil shift

As an atom absorbs a photon, conservation of momentum increases the atom momentum by the photon momentum  $\Delta p = \hbar k$ , where  $k = 2\pi/\lambda$  is the wavenumber. As a consequence the kinetic energy of the atom increases by  $\Delta E = \Delta p^2/2m = \hbar^2 k^2/2m \approx h \times 63.5$  kHz, and the absolute transition frequency should be corrected for this energy shift. Note that the accuracy in the recoil shift correction is limited by the accuracy of the measured transition frequency (~ 9 digits accuracy) and known atomic mass (12 digits accuracy [88]) and therefore does not contribute to the error budget.

<sup>&</sup>lt;sup>1</sup>With the trap frequencies known, the size of the thermal cloud is simply  $x_i = \sqrt{k_B T/m\omega_i^2}$ , and for the Bose-Einstein condensate with known chemical potential  $\mu$  the size is  $x_i = \sqrt{2\mu/m\omega_i^2}$ . In the harmonic approximation this allows a quick estimate of the spatial variation of the ac Stark shift, which is below 50 kHz for the most extreme experimental conditions (i.e. cloud temperature, atom numbers and ODT powers).

**Table 3.1** – Polarizabilities of the 2  ${}^{3}S_{1}$  and 2  ${}^{1}P_{1}$  states at 886.6 nm and 1557.3 nm calculated using Eq. 2.5 and summing over all transitions up to n = 10. The ac Stark shift contributions are estimated for our experimental parameters (beam waists and powers).

Wavelength [nm]	$\alpha(2 \ ^1P_1) \ [a_0^3]$	$\alpha(2 \ {}^3S_1) \ [a_0^3]$	ac Stark shift [kHz]
886.6	$4.1  imes 10^2$	-60	-0.3
1557.3	$-1.0  imes 10^2$	$6.0  imes 10^2$	30

#### ac Stark shift

Ab initio calculations of the polarizabilities of both the 2  ${}^{3}S_{1}$  and 2  ${}^{1}P_{1}$ states at both the wavelength of the ODT and the spectroscopy beam are combined with previously performed ac Stark shift measurements [31] as a cross-check. The calculations are based on a simplified model based on Eq. 2.5 by summing over all transitions up to n = 10 states which has shown to be within 1% of the extended ac Stark shift calculations [133] and is therefore more than sufficient for our purposes. The polarizabilities are given in Table 3.1 together with the estimated ac Stark shifts, based on estimates for the power and beam waists of both the ODT and the spectroscopy beam. The absolute accuracy on these estimates is on the order of 10 kHz due to uncertainties in the calculation of the peak intensity of the ODT, but are negligible compared to the statistical accuracy of the frequency determination of ~ 0.5 MHz.

#### Zeeman shift

Using the rf resonance method as explained in Chapter 2, the Zeeman shift is measured a few times during a single day with kHz precision and on the order of  $\Delta E \approx h \times 1.5$  MHz. The individual linescans are corrected (on a daily basis) with an interpolated value of the Zeeman shift which is accurate to a few kHz.

Apart from a direct correction of the Zeeman shift, we also consider the possibility of exciting two transitions ( $\pi$  and  $\sigma^{-}$ ) and their effect on any possible line splitting. For this we calculate the relative transition strengths based on the geometry of our experiment. The Zeeman energy shift of an atomic state is  $\Delta E = m_J g_J \mu_B B$ , where  $\mu_B$  is the Bohr magneton,  $g_J$  the Landé factor of the fine structure state,  $m_J$  the quantum number of the fine structure magnetic state and B the magnitude of the local magnetic field. The Landé factor is given as [114]

$$g_J = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}.$$
(3.8)

The initial spin-stretched state is 2  ${}^{3}S_{1}$   $m_{J} = +1$ . With an undefined polarization of the driving field there are two possible transitions: either a  $\pi$  ( $\Delta m = 0$ ) transition to the 2  ${}^{1}P_{1}$   $m_{J} = +1$  state or a  $\sigma^{-}$  ( $\Delta m = -1$ ) transition to the 2  ${}^{1}P_{1}$   $m_{J} = 0$  state. Filling in the corresponding quantum numbers, we find a Landé factor of  $g_{J} = 2$  for the 2  ${}^{3}S_{1}$  and  $g_{J} = 1$  for the 2  ${}^{1}P_{1}$  state. The differential shift of the  $\pi$  and  $\sigma^{-}$ transitions are therefore

$$\Delta_{\pi} = -\mu_B B, \qquad \pi\text{-transition}, \tag{3.9}$$

$$\Delta_{\sigma} = -2\mu_B B, \quad \sigma^{-} \text{-transition.} \tag{3.10}$$

The population of the excited state is proportional to the square of the transition dipole matrix element  $\mu_{eg}$ , which is defined in the  $|Jm_J\rangle$  basis as [114]

$$\mu_{eg} = e \langle J' \, m'_J | \vec{\varepsilon} \cdot \vec{r} | J \, m_J \rangle, \qquad (3.11)$$

where  $\vec{\varepsilon}$  is the polarization unit vector and  $\vec{r}$  the position vector. The transition dipole matrix element can be decomposed in the  $|L m_L, S m_S\rangle$  basis using the appropriate Clebsch-Gordan coefficients such that

$$\mu_{eg} = e(-1)^{L'+S-m'_J} \sqrt{(2J+1)(2J'+1)} \\ \times \begin{cases} L' & J' & S \\ J & L & 1 \end{cases} \begin{pmatrix} J & 1 & J' \\ m_J & q & -m'_J \end{pmatrix} \langle \beta' L' | r | \beta L \rangle, \quad (3.12)$$

where the term in the curly brackets is a 6j symbol, the term in the round brackets is a 3j symbol, and  $\beta$ ,  $\beta'$  denote the remaining quantum numbers that are not used in this calculation. The information about the polarization of the light (and the induced transition) is now given by the factor q in the 3j symbol. Here  $q = 0, \pm 1$  depending on the

induced  $\pi$ , or  $\sigma^{\pm}$  transition. Filling in all relevant quantum numbers for the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition we obtain

$$\mu_{eg}^{\pi} = \mu_{eg}^{\sigma^{-}} = -\frac{e}{\sqrt{6}} \langle \alpha' L' | r | \alpha L \rangle.$$
(3.13)

The polarization of the spectroscopy light in our experiment is linear, but the k-vector of the light is incident on the quantization axis of the B-field with angle  $\theta$ . This means that the linear polarization unit vector  $\vec{\varepsilon}$  (see Eqn. 3.11) should be decomposed in a parallel and transverse component, as in

$$\vec{\varepsilon} = \sin\theta \cdot \vec{\varepsilon}_{\parallel} + \cos\theta \cdot \vec{\varepsilon}_{\perp}. \tag{3.14}$$

The parallel component induces  $\pi$ -transitions and the transverse component induces  $\sigma^{\pm}$ -transitions. Therefore the vector  $\vec{\varepsilon}_{\perp}$  should be decomposed into a superposition of left- and right-handed circularly polarized light

$$\vec{\varepsilon}_{\perp} = \frac{1}{\sqrt{2}} \left( \vec{\varepsilon}_{+} + \vec{\varepsilon}_{-} \right). \tag{3.15}$$

This gives us the total decomposition of the incoming polarization into three components which will induce the three transitions as

$$\vec{\varepsilon} = \sin\theta \cdot \vec{\varepsilon}_{\parallel} + \frac{1}{\sqrt{2}}\cos\theta \cdot \vec{\varepsilon}_{+} + \frac{1}{\sqrt{2}}\cos\theta \cdot \vec{\varepsilon}_{-}.$$
 (3.16)

Noting from Eqn. 3.11 that  $\mu_{eg} \propto \vec{\varepsilon}$ , the effective transition dipole matrix elements weighed by the polarization components give

$$\mu_{\rm eff}^{\pi} = \sin\theta \cdot \mu_{eg}^{\pi} = -\sin\theta \cdot \frac{e}{\sqrt{6}} \langle \alpha' L' | r | \alpha L \rangle, \qquad (3.17)$$

$$\mu_{\text{eff}}^{\sigma^{-}} = \frac{1}{\sqrt{2}} \cos \theta \cdot \mu_{eg}^{\sigma^{-}} = -\cos \theta \cdot \frac{e}{\sqrt{12}} \langle \alpha' L' | r | \alpha L \rangle.$$
(3.18)

As the transition strength  $S \propto \mu_{\text{eff}}^2$  and from the geometry of the setup we find  $\sin \theta \approx 0.1$ , the ratio of the transition strengths of the  $\pi$ - and  $\sigma^-$ -transitions is

$$\frac{S^{\pi}}{S^{\sigma^-}} = 2 \frac{\sin^2 \theta}{\cos^2 \theta} \approx 0.02.$$
(3.19)

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This means that the contribution of  $\pi$ -transitions is only significant at the percent level. A simple simulation of a dual-peak structure with this amplitude ratio and a differential Zeeman splitting of  $\Delta_{\pi} - \Delta_{\sigma} = \mu_B B \approx$  $0.7 \text{ MHz} = \Gamma/500$  shows that there is a systematic shift of  $\sim 14 \text{ kHz}$  due to this effect. Although it is necessary to take this effect into account, again, it is much smaller than the statistical accuracy of the frequency determination.

#### Mean field shift

The mean field shift, also known as the cold-collision shift, is caused by the fact that two colliding  $2 {}^{3}S_{1}$  atoms have a different *s*-wave scattering length than a  $2 {}^{3}S_{1}$  atom colliding with a  $2 {}^{1}P_{1}$  atom. This difference in scattering lengths leads to a different chemical potential for a gas of purely  $2 {}^{3}S_{1}$  atoms and a mixture of  $2 {}^{3}S_{1}$  and  $2 {}^{1}P_{1}$  atoms as the chemical potential of one component is  $\mu_{i} = (4\pi\hbar^{2}/m)(g_{ii}^{(2)}a_{ii}n_{i} + g_{ij}^{(2)}a_{ij}n_{j})$ , where  $a_{ij}$  is the respective scattering length,  $n_{i}$  the atom number density and  $g_{ij}^{(2)}$  the second order correlation function at zero separation distance<sup>2</sup>. In the limit of much less  $2 {}^{1}P_{1}$  atoms than  $2 {}^{3}S_{1}$  atoms the mean field shift is approximated as

$$h\Delta\nu = \delta\mu = \frac{4\pi\hbar^2}{m}n_S(a_{SS} - a_{SP}).$$
(3.20)

This expression could already be used to make a rough estimate of the mean field shift as function of the unknown  $a_{SP}$  scattering length, but it is also obvious that this shift diverges for  $a_{SP} \to \infty$  which is not a realistic expectation. To solve this issue we need to include the collision energy in the description of the mean field shift.

The actual cold-collision cross-section  $\sigma$  is not only a function of the scattering length *a* between two atoms, but also of the collision energy that is described by the relative collision momentum *k*. The relative momentum *k* is defined such that the total kinetic energy of the collision is  $E_{kin} = \hbar^2 k^2/2\mu$ , with  $\mu$  the reduced mass of the system. To first order

<sup>&</sup>lt;sup>2</sup>The second order correlation function  $g^{(2)}(0) = 1$  for coherent bosons (i.e. in a Bose-Einstein condensate) and  $g^{(2)}(0) = 2$  for a thermal Bose gas [259].

in k, the cross-section is

$$\sigma = \frac{4\pi a^2}{1 + k^2 a^2}.\tag{3.21}$$

In the limit of very small collision energy or scattering length,  $ka \ll 1$ , we obtain the hard-sphere scattering result  $\sigma_0 = 4\pi a^2$ . In the other limiting case,  $ka \gg 1$ , the collision cross-section is bound as  $\sigma(k) = 4\pi/k^2$ . This is known as the unitarity limit where the scattering length is so large that the collision cross-section only depends on the collision energy.

Without any knowledge about the 2  ${}^{3}S_{1} - 2 {}^{1}P_{1}$  molecular potential, there is no *a priori* indication of the sign and magnitude of the 2  ${}^{3}S_{1} - 2 {}^{1}P_{1}$  scattering length  $a_{SP}$ . A suitable model has been used by Kokkelmans *et al.* to calculate the mean field shift and line broadening of a rubidium clock transition [260]. The mean field shift  $\delta \omega$  and line broadening  $\gamma$  of the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition can be described by a sum over all relevant atomic states j as

$$i\delta\omega - \gamma = \sum_{j} n_j \langle v(i\lambda_j - \sigma_j) \rangle,$$
 (3.22)

with  $\langle ... \rangle$  defining a thermal average,  $n_j$  the density of atoms in state j, v the relative velocity between two colliding particles and  $\lambda_j$  and  $\sigma_j$  the shift and width cross-sections. These cross-sections can be obtained from the S-matrices of the collisions as

$$i\lambda_j - \sigma_j = (1 + \delta_{1j})(1 + \delta_{2j})\frac{\pi}{k^2} \times \sum_l (2l+1)[S^l_{(1j),(1j)}S^{l\star}_{(2j),(2j)} - 1],$$
(3.23)

with l the partial wave index. In the s-wave collision regime we can limit ourselves to l = 0 and the S-matrices become

$$S_{(nj),(nj)} = \frac{1 - ika_{nj}}{1 + ika_{nj}}.$$
(3.24)

For the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition, the density of excited state atoms can be ignored due to the fast decay to the 1  ${}^{1}S_{0}$  ground state, and the

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**Figure 3.7** – Mean field shift calculated for any scattering length  $a_{SP}$  in the range  $-10^4 a_0 < a_{SP} < 10^4 a_0$ . The shaded area is the uncertainty in the mean field shift due to the uncertainty in the average density as given in the text. The maximum shift is  $\pm 90$  (20) kHz at scattering lengths of 2700  $a_0$  or  $-2400 a_0$ .

summation over the states is reduced to a complex function  $\Omega$  defined as

$$\Omega \equiv i\delta\omega - \gamma = \frac{4\pi\hbar}{mk} n_S \Big( \frac{1 - ika_{SS}}{1 + ika_{SS}} \cdot \frac{1 + ika_{SP}}{1 - ika_{SP}} - 1 \Big). \tag{3.25}$$

Here  $a_{SS}$  is the 2  ${}^{3}S_{1} - 2 {}^{3}S_{1}$  scattering length of the  ${}^{5}\Sigma_{g}^{+}$  potential and is  $a_{SS} = 7.512(5)$  nm = 142.0(1)  $a_{0}$ , where  $a_{0}$  is the Bohr radius [152]. From this final expression we can find the broadening and shift as  $\gamma = -\text{Re}(\Omega)$  and  $\delta\omega = \text{Im}(\Omega)$ .

The kinetic energy of the atoms in the BEC can be estimated from the width of the momentum distribution of the atoms in the BEC [203] and we find an energy corresponding to a few nK. For the thermal atoms the temperature is 0.2  $\mu$ K in our experiments. The absorption of a 887-nm photon will increase the kinetic energy of a helium atom by about 3  $\mu$ K which is much larger than any of the kinetic energies of the thermal and condensed atoms. Therefore we ignore any initial velocity distribution



**Figure 3.8** – Broadening calculated for any scattering length  $a_{SP}$  in the range  $-10^4 a_0 < a_{SP} < 10^4 a_0$ , where the shaded area is the uncertainty in the broadening due to the uncertainty in the average density as given in the text.

of the gas and describe the collision energy solely by the recoil energy of the 887-nm photon.

Using these parameters, and an average peak density of the BEC of  $2.0(0.4) \times 10^{13}$  cm<sup>-3</sup>, the mean field shift and broadening can be calculated for any possible value of  $a_{SP}$ . The results are shown in Figures 3.7 and 3.8. We find the maximum possible mean field shift to be  $\pm 90(20)$  kHz at scattering lengths  $a_{SP} = 2700 \ a_0$  or  $a_{SP} = -2400 \ a_0$ . Furthermore, the maximum broadening is  $\leq 200$  kHz, which is negligible compared to the natural linewidth of 287 MHz.

Although the shift represents almost 20% of the statistical error bar on our determined transition frequency, the scattering lengths at which this would occur are very large. In order to 'accidentally' have such a large scattering length, the collision energy should be resonant with a bound state in the  $2 {}^{3}S_{1} - 2 {}^{1}P_{1}$  molecular potential. The typical energy spacing of such bound states is on the order to 10's of GHz, and with a linewidth of approximately 287 MHz the probability of being resonant is at the percent level and therefore quite unlikely. Furthermore, the used



**Figure 3.9** – Example of a single linescan measurement, showing 90 consecutive measurements over a range of approximately 1.8 GHz. A Lorentzian fit is used to determine the transition frequency and the linewidth. The residuals are shown in the lower plot. The frequency axis is centered on the transition frequency determined in this scan.

model does not incorporate the short lifetime of 0.55 ns of the excited state. The finite lifetime would reduce the mean field shift even more, just as any Penning ionization processes that could occur between these states [261]. These last two arguments allow us to assume that the mean field shift is much less than  $\pm 90$  kHz and therefore negligible compared to the uncertainty in our determined transition frequency.

# 3.5 Results

An example of a single normalized line scan of the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$ transition is shown in Figure 3.9. The scan spans a range of 1.8 GHz measured at a 20 MHz interval and would take about 45 minutes to acquire. The line is fit with a Lorentzian line shape, and from the residuals shown in Figure 3.9 we see that this is indeed an excellent fit. In a period of two months in summer 2013 over 100 line scans were acquired of which 77 remained, as some scans were discarded due to a



**Figure 3.10** – Determined transition frequency averaged per measurement day, based on a total of 77 measurements. The error bars on the data represent the  $1\sigma$  standard deviation of the daily average. The frequencies are centered around the final average transition frequency and the gray bar represents its  $1\sigma$  standard deviation of 0.5 MHz.

strong variation in the atom number loading or because the spectroscopy laser went out of lock partially through the measurement. From these measurements we can determine the absolute transition frequency to determine the ionization energy and lifetime of the 2  ${}^{1}P_{1}$  state.

#### $2 {}^{1}P_{1}$ ionization energy

Based on a total of 77 line scans, the daily average transition frequency is shown in Figure 3.10. We obtain a  $2 {}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition frequency of 338 133 594.4 (0.5) MHz. This value is in good agreement with the most recent theoretical value of 338 133 594.9 (2.7) MHz [26], where the accuracy is limited by the QED calculations of the 2  ${}^{3}S_{1}$  state.

From our previously measured 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$  transition frequency (192510702.1456 (0.0018) MHz [31]) we extract a 2  ${}^{1}S_{0} \rightarrow 2 {}^{1}P_{1}$  transition frequency of 145622892.2 (0.5) MHz. This result agrees with the recent 2  ${}^{1}S_{0} \rightarrow 2 {}^{1}P_{1}$  frequency measurement [33] within 0.6 (0.6) MHz. The 2  ${}^{3}S_{1}$  IE of 1152842742.97 (0.06) MHz, derived from a measure-



**Figure 3.11** – Comparison of our experimental result for the 2  ${}^{1}P_{1}$  IE with other experiments [33, 34, 249] and QED theory by Yerokhin and Pachucki [26]. All recent experimental results show  $a > 3\sigma$  discrepancy with theory.

ment of the 2  ${}^{3}S_{1} \rightarrow 3 {}^{3}D_{1}$  transition frequency [32] and the calculated 3  ${}^{3}D_{1}$  IE [52], can now be combined with our result to give a 2  ${}^{1}P_{1}$  IE of 814 709 148.6 (0.5) MHz. Comparing this result to both measurements of Luo *et al.* [33, 34], we find very good agreement. An overview of the most accurate experimental results and the QED calculations for the 2  ${}^{1}P_{1}$  IE is shown in Figure 3.11. A discrepancy of > 3 $\sigma$  with the theoretical IE as calculated by Yerokhin and Pachucki [26] remains. As QED calculations of most low-lying states of <sup>4</sup>He<sup>\*</sup> agree very well with experiment, improved calculations for the 2  ${}^{1}P_{1}$  state are now timely. It may be that the contribution of  $m\alpha^{7}$  terms is not treated well in this case, as a re-evaluation of these terms has shifted the IE by almost 1 MHz [26, 55].

# $2 {}^{1}P_{1}$ lifetime

Based on the same 77 linescans from which we determine the transition frequency we find a natural linewidth of  $289(2)_{\text{stat}} \binom{+0}{-7}_{\text{syst}}$  MHz. This



**Figure 3.12** – Previously experimentally determined 2  ${}^{1}P_{1}$  lifetimes [251–255] compared to our result and the theoretical result by Morton, Moffat and Drake [250]. Our result contains an extended, dashed, error bar indicating a systematic uncertainty as discussed in the text.

includes the line shape and broadening corrections as discussed in Section 3.3, and corresponds to a lifetime of the 2  ${}^{1}P_{1}$  state of  $0.551(0.004)_{\text{stat}} ({}^{+0.013}_{-0.000})_{\text{syst}}$  ns. This result is shown in Figure 3.12 together with previously determined lifetimes [251–255] and shows an improvement in the accuracy compared to the previous most accurate determination. Our result is in agreement with the previous measurements, which are all based on completely different techniques, and agrees with a theoretical lifetime of 0.5555 ns, which is accurate to the last digit and calculated neglecting finite mass and relativistic effects that are expected below the 0.1% accuracy level [250].

The systematic uncertainty included in this result is based on possible saturation of the MCP detector, which can lead to broadening of the line shape. Saturation effects can be expected and have been observed in other experiments with metastable helium BECs [125, 162], and we investigate any possible nonlinear behavior of the MCP detector by analyzing the determined linewidths as function of the average loaded atom number with a quadratic function. From the fits we de-



Figure 3.13 – Experimentally determined linewidths as a function of the total number of atoms loaded into the ODT. The horizontal line represents the statistical average of 289(2) MHz. The dotted line is a quadratic function as mentioned in the text, which gives a natural linewidth of 282(5) MHz and a quadratic term coefficient of  $5(6) \times 10^{-13}$  MHz/atoms<sup>2</sup>.

termine a quadratic coefficient  $5(6) \times 10^{-13}$  MHz/atoms<sup>2</sup>, as shown in Figure 3.13 and the determined natural linewidth shifts by -7 MHz from 289(2) MHz to 282(5) MHz. Although this is barely significant, a systematic uncertainty of -7 MHz is added to the result to indicate the worst-case shift in the linewidth if we allow a nonlinear response of the MCP detector in our analysis.

### 3.6 Conclusion

To summarize, we have measured the  $2 {}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition frequency in a quantum degenerate gas of  ${}^{4}\text{He}^{*}$  to  $1.6 \times 10^{-9}$  relative accuracy. From this measurement the  $2 {}^{1}P_{1}$  IE is determined with  $6.7 \times 10^{-10}$ relative accuracy, in agreement with two recent independent determinations by Luo *et al.* [33, 34]. We show a >  $3\sigma$  discrepancy in the  $2 {}^{1}P_{1}$  IE with the most accurate QED calculation by Yerokhin and Pachucki [26],
indicating that a renewed effort on the QED calculations is required. We also report the most accurate determination of the 2  ${}^{1}P_{1}$  lifetime to date. This new determination is in agreement with theory and all previous experimental determinations.

# CHAPTER 4

### Line shapes of the $2 \ {}^{3}S \rightarrow 2 \ {}^{1}S$ transition for quantum degenerate bosons and fermions

#### Abstract

We observe a dramatic difference in optical line shapes of a <sup>4</sup>He Bose-Einstein condensate and a <sup>3</sup>He degenerate Fermi gas by measuring the 1557-nm 2 <sup>3</sup>S - 2 <sup>1</sup>S magnetic dipole transition (8 Hz natural linewidth) in an optical dipole trap. The 15 kHz FWHM condensate line shape is only broadened by mean field interactions, whereas the degenerate Fermi gas line shape is broadened to 75 kHz FWHM due to the effect of Pauli exclusion on the spatial and momentum distributions. The asymmetric optical line shapes are observed in excellent agreement with line shape models for the quantum degenerate gases. For <sup>4</sup>He a triplet-singlet s-wave scattering length  $a = +50(10)_{\text{stat}}(43)_{\text{syst}} a_0$  is extracted.

The high spectral resolution reveals a doublet in the absorption spectrum of the BEC, and this effect is understood by the presence of a weak optical lattice in which a degeneracy of the lattice recoil and the spectroscopy photon recoil leads to Bragg-like scattering.

This chapter is based on: Comparison of spectral linewidths for quantum degenerate bosons and fermions, R.P.M.J.W. Notermans, R.J. Rengelink, and W. Vassen, Physical Review Letters **117**, 213001 (2016)

#### 4.1 Introduction

The bosonic or fermionic nature of a particle is a fundamental property. and trapped quantum degenerate gases display dramatic different behavior depending on the quantum statistical nature of the gas. At low temperatures identical bosons accumulate in the lowest state in the trap. leading to Bose-Einstein condensation. In contrast, identical fermions cannot occupy the same state due to the Pauli exclusion principle, and will 'fill' all states in the trap from the bottom up until no more atoms - or states - are available. A drastic difference in line shape of a narrow optical transition is expected when measured in a Bose-Einstein condensate (BEC) and a degenerate Fermi gas (DFG). In this work we show a direct comparison of this difference between a BEC of metastable <sup>4</sup>He and a DFG of metastable <sup>3</sup>He trapped in an optical dipole trap (ODT). We do this work in the framework of high-precision frequency metrology in helium, aimed at testing quantum electrodynamics (QED). Comparison of accurate transition frequencies is used to determine fundamental physical parameters that are difficult to measure otherwise, such as the nuclear charge radius of an atom. Recently high-precision frequency metrology in (muonic) hydrogen and deuterium resulted in a remarkable discrepancy in the determination of the proton and deuteron charge radius [95, 96]. This discrepancy, also known as the 'proton radius puzzle', is currently under scrutiny by many groups all over the world and similar work is ongoing for helium [99, 100]. To determine the  ${}^{3}\text{He}{}^{4}\text{He}$ nuclear charge radius difference, we recently measured the doubly forbidden 2  ${}^{3}S - 2 {}^{1}S$  transition at 1557 nm (natural linewidth 8 Hz) in both quantum degenerate  ${}^{4}$ He and  ${}^{3}$ He with 1.8 kHz and 1.5 kHz accuracy, respectively [31]. The measured isotope shift, combined with QED calculations, allowed a determination of a squared nuclear charge radius difference of 1.028(11) fm<sup>2</sup> [27]. To compare this determination to measurements in muonic helium ions [99, 100] we aim to measure the 2  ${}^{3}S - 2 {}^{1}S$  transition frequency with  $\ll 1$  kHz accuracy. Using a narrow linewidth spectroscopy laser we are able to observe asymmetric line shapes for a BEC and a DFG of metastable helium as well as a line splitting in the optical spectrum of the BEC. Quantification of these effects by understanding the line shapes is essential in achieving the sub-kHz accuracy goal.

#### 4.2 Experimental setup

Our experimental setup is similar to earlier work [31] and to a more recent measurement of the  $2^{3}S_{1} - 2^{1}P_{1}$  transition at 887 nm [35]. We load a BEC of typically 10<sup>6</sup> atoms in the metastable 2  ${}^{3}S_{1}$   $(m_{J} = +1)$ state (lifetime  $\sim 7800$  s, internal energy 19.82 eV) into a crossed-beam ODT operating at 1557.3 nm. The crossing angle between the ODT beams is  $19^{\circ}$ , and the temperature of the thermal atoms in the ODT is typically  $T \approx 0.2 \ \mu \text{K}$ . As the fermionic <sup>3</sup>He atoms cannot thermalize once their temperature is below the p-wave barrier, they are loaded simultaneously with <sup>4</sup>He and sympathetically cooled to degeneracy [131]. The quantum degenerate <sup>3</sup>He-<sup>4</sup>He mixture is loaded into the ODT to rethermalize, after which the <sup>4</sup>He atoms are blown away using resonant light. This procedure leaves a pure DFG of thermalized <sup>3</sup>He in the 2  ${}^{3}S_{1}$   $(F = \frac{3}{2}, m_{F} = +\frac{3}{2})$  state. The spectroscopy beam copropagates with one of the ODT beams in order to overlap with the trapped cloud. The atoms are probed for a few seconds, after which the remaining cloud is released from the ODT. The time-of-flight signal of the metastable atoms is measured on a microchannel plate (MCP) detector and used to determine the remaining atom number, temperature and chemical potential. The measurements alternate with and without the spectroscopy light in order to have a continuous background measurement to normalize the line shapes.

For this experiment a narrow linewidth fiber laser is transfer-locked to an ultrastable (< 2 Hz) laser system operating at 1542 nm using a caesium clock-referenced femtosecond frequency comb to bridge the 15 nm wavelength difference between both lasers. Due to uncompensated fiber links we estimate a residual ~ 5 kHz linewidth of the spectroscopy laser, which is in agreement with the 4.5 kHz linewidth (FWHM) determined in our line shape fits (see Appendix 4.D). This is a factor 20 improvement compared to our previous experiment [31].

## 4.3 Comparing the line shape of a BEC to a degenerate Fermi gas

Figure 4.1 shows the optical  $\sigma^-$  transitions measured in a BEC [2  ${}^3S_1$  $(m_J = +1) \rightarrow 2 \, {}^{1}S_0 \, (m_J = 0)]$  and DFG  $[2 \, {}^{3}S_1 \, (F = \frac{3}{2}, m_F = +\frac{3}{2}) \rightarrow 2 \, {}^{1}S_0 \, (F = \frac{1}{2}, m_F = +\frac{1}{2})]$ . The uncertainty in the frequency is 1.8 kHz, and the error bars in the normalized atom numbers are based on the atom number fluctuations in the measurements. The zero on the frequency axis represents the transition frequency from the bottom of the trap which is not measured as an absolute frequency. For the DFG results the atom number  $N \approx 3 \times 10^5$  and peak density  $\sim 1 \times 10^{12}$  cm<sup>-3</sup>. There are three times as many atoms in the BEC compared to the DFG due to the more complicated loading procedure of the DFG [131], and the peak density of the BEC is ten times higher. Despite this, the line shape of the DFG is over five times broader. This is caused entirely by the broad momentum and spatial distribution of the fermions. In contrast, the BEC line shape only has a finite width due to the mean field interactions (which are absent in a coherent excitation of a Fermi gas [262]) and the linewidth of the spectroscopy laser. Without the effects of quantum statistics the width of both line shapes would simply be the Doppler width (31 kHz for <sup>4</sup>He, 35 kHz for <sup>3</sup>He). This huge difference in linewidths based on the quantum statistics of the helium isotopes is complementary to the observation of bunching and antibunching with the same atoms [163]. For frequency metrology purposes it is clear that proper modeling is imperative in order to determine the true transition frequency.

The line shape for the DFG is calculated using the absorption line profile from Ref. [263] and involves explicit integration of the Fermi-Dirac distribution of the spatial and momentum states occupied in the ODT, convolved with a Lorentzian distribution with a FWHM of 4.5 kHz (determined from the BEC fits) to model the finite linewidth of the spectroscopy laser. Time-dependent depletion of the DFG does not play a role because the fermions neither rethermalize nor redistribute over the trap states during the optical excitation. Using the experimentally determined degeneracy  $T/T_F = 0.33(7)$  and chemical potential  $\mu/k_B = 0.55(15) \ \mu\text{K}$  of the DFG, the calculated line shape is shown in



**Figure 4.1** – Direct comparison of the (normalized) optical line shapes of the  $2 {}^{3}S - 2 {}^{1}S$  transition measured in a degenerate Fermi gas (top) and a Bose-Einstein condensate (bottom) of metastable helium. The full lines represent the fits provided by the models discussed in the main text, and display a small but significant asymmetry. For a clear comparison only one peak of the observed BEC doublet is shown (see Figure 4.4). The zero frequency represents the transition frequency from the bottom of the trap.

Figure 4.1 (top). As only the relative amplitude and frequency offset of the line are fitted to the data, the model predicts the line shape perfectly. Although hardly visible, the line shape is asymmetric and the model provides a reduced  $\chi^2 = 1.09$ .

The line shape for light absorption from a BEC is fundamentally different from that of a DFG and was first calculated by Killian for the absorption on the 1S - 2S transition in a hydrogen BEC [264, 265]. Excellent agreement with the data was demonstrated, but the line shape function [258] cannot be used in our experiment for two reasons. First, in [258] it is assumed that the trapping potentials of both the initial and final state are equal. This assumption is invalid in our ODT as the ratio of the polarizabilities of both states  $\alpha_s/\alpha_t = -1.64(1)$  (see Chapter 5 or Ref. [133]), where s and t denote the singlet and triplet state  $(2^{1}S \text{ atoms})$ are repelled from the trap). Second, the excitation fraction in [258] was on the order of 1% and therefore depletion of the condensate during excitation could be neglected. This is invalid in our experiment as the excited BEC fraction is typically 20 - 70% to have an acceptable signalto-noise ratio. Therefore we extend the Killian model [258], of which the full derivation is given in Appendix 4.A, by including the polarizabilities in the effective potentials of the initial and final state. This results in the addition of the ac Stark shift to the resonance condition, and an effective rescaling of the mean field shift term  $(4\pi\hbar^2 n_0/m)(a_{ts}-a_{tt})$  which becomes  $(4\pi\hbar^2 n_0/m)(a_{ts} - (\alpha_s/\alpha_t)a_{tt})$ . Here  $n_0$  is the peak density of the condensate,  $a_{tt}$  the 2  ${}^{3}S_{1} - 2 {}^{3}S_{1}$  s-wave scattering length in the pure  ${}^{5}\Sigma_{a}^{+}$  potential, and  $a_{ts}$  the 2  ${}^{3}S_{1}(m_{J}=+1)-2$   ${}^{1}S_{0}(m_{J}=0)$  s-wave scattering length. Although  $a_{ts}$  has not been measured or calculated to date,  $a_{tt}$  is very accurately known:  $a_{tt}^{\text{theory}} = 143.0(5) a_0 [144, 146]$  and  $a_{tt}^{exp} = 142.0(1) a_0$  [152], where  $a_0$  is the Bohr radius.

It is convenient to express the line shape of the BEC using the chemical potential  $\mu = 4\pi \hbar^2 a_{tt} n_0/m$ , which we determine directly from a time-of-flight measurement. The line shape  $S(\nu, \mu)$ , derived in Appendix 4.A, is

$$S(\nu,\mu) = \frac{15\pi\hbar\Omega_R^2}{4}N\frac{h\nu}{\tilde{\mu}^2}\sqrt{1-\frac{h\nu}{\tilde{\mu}}},\tag{4.1}$$

where  $\Omega_R$  is the Rabi frequency of the transition, N the total atom number of the BEC,  $\nu$  the detuning from the absolute transition frequency including the full ac Stark shift of the trap, and  $\tilde{\mu} = (a_{ts}/a_{tt} - \alpha_s/\alpha_t)\mu$  the rescaled chemical potential of the BEC (see Section 4.A). This rescaling shows how the mean field interaction and ac Stark shift affect the effective potential experienced by the atoms. The line shape of the BEC is asymmetric with a high-frequency cut-off at  $\nu = 0$ .

As the atom number of the condensate scales as  $N \propto \mu^{5/2}$  in the Thomas-Fermi limit and the line shape  $S(\nu, \mu)$  constitutes a one-body loss process, the decay of the chemical potential of the BEC during the spectroscopy phase can be written as

$$\frac{d\mu}{dt} = \frac{2}{5} \frac{\mu}{N} \tilde{S}(\nu, \mu) - \frac{2}{5} \Gamma \mu, \qquad (4.2)$$

where  $\tilde{S}(\nu,\mu)$  is the line shape  $S(\nu,\mu)$  convolved with a Lorentzian distribution to model the spectroscopy laser linewidth (as derived in Appendix 4.A). We include the one-body lifetime  $\Gamma^{-1}$  of the gas as the typical interaction times are long enough (1-6 seconds) that one-body loss cannot be neglected. The decay of the chemical potential is slow enough such that the condensate can be assumed to remain in equilibrium throughout the excitation. The BEC is held in the ODT for 4-5 seconds before switching on the probe light so two- and three-body loss processes are negligible. The nonlinear differential Eq. 4.2 is numerically solved to fit to the line shape as shown in Figure 4.1 (bottom). Here we use only the frequency offset and  $a_{ts}$  scattering length as free parameters, giving a reduced  $\chi^2 = 0.94$ .

#### 4.4 Bragg-like scattering in an optical lattice

Interestingly, we observe a doublet in the BEC spectrum where a single peak was expected. This double peak structure is attributed to the presence of a weak optical lattice in our crossed dipole trap due to birefringence in our vacuum windows. The ODT laser wavelength  $\lambda_{\text{ODT}} \approx 1557.3$  nm (sufficiently off-resonance from the  $2^{3}S - 2^{1}S$  transition to have negligible scattering) is close to the transition wavelength and creates a lattice with periodicity  $d = \lambda_{\text{ODT}}/[2\cos(\theta/2)]$  and effective lattice recoil energy  $E_{r}^{l} = \hbar^{2}q_{l}^{2}/2m$ , where  $q_{l} = \pi/d$ . This recoil energy is nearly degenerate with the recoil when absorbing a spectroscopy



Figure 4.2 – (a) In the crossed-beam optical dipole trap geometry we have a weak optical lattice with periodicity  $d = \lambda_{ODT}/[2\cos(\theta/2)]$ . Absorption of a spectroscopy photon results in a recoil momentum  $q' = 2\pi\cos(\theta/2)/\lambda_{spec}$  in the lattice direction equal to the lattice recoil momentum  $\pi/d$ . (b) Band structure of the optical lattice for the ground (lower red bands) and upper state (upper blue bands) for a typical lattice amplitude  $V_0 \approx 2E_r^l$ . The BEC is situated at quasimomentum q = 0 in the lowest band (black dot). Absorption of a photon creates a quasimomentum  $q' \approx \pi/d$  in the optical lattice for the excited state. Absorption can take place if the spectroscopy laser frequency is resonant with the lowest or first band at the edge of the Brillouin zone (black dots at  $q = \pi/d$ ), giving rise to the observed bandgap splitting  $\Delta$ . 112

photon in the lattice frame,  $E_r = \hbar^2 q'^2/2m \approx h \times 20.0$  kHz, with  $q' = 2\pi \cos(\theta/2)/\lambda_{\rm spec}$ , see Figure 4.2(a). The absorbed spectroscopy photon provides the excited wavefunction a quasimomentum q' in the frame of the lattice. This quasimomentum is at the edge of the first Brillouin zone and therefore at the optical lattice bandgap, as shown in Figure 4.2(b). The resonance condition can only be satisfied below or above the bandgap, leading to a line splitting  $\Delta$  of the transition, where  $\Delta = V_0/2$  and  $V_0$  is the optical lattice modulation amplitude as observed by the excited state [121, 197].

This excitation in a weak optical lattice is reminiscient of Bragg scattering of a BEC in an applied optical lattice [266, 267]. Contrary to Bragg scattering, where an applied moving optical lattice causes diffraction, the direct one-photon optical excitation causes the transition to a higher momentum state near the edge of the Brillouin zone. We verify the presence of the weak optical lattice by rotating the polarization of the second ODT beam with respect to the first. Figure 4.3 shows that the splitting increases as  $V_0$  is increased and in these measurements we estimate the optical lattice modulation amplitude for the 2  $^{1}S$  state to be  $V_0 \leq 6.5 E_r^l$  for the largest splitting shown. As the polarizability for the 2  ${}^{3}S$  atoms is smaller by a factor 1.64, the optical lattice observed by the BEC is  $V_0 \leq 4.0E_r^l$  for the largest splitting and the ultracold cloud is in the superfluid regime [268]. Aspect ratio inversion in absorption images of the expanding cloud confirms this. In this regime the mean field description is applicable and coupling to higher lattice bands can be ignored. The doublet is simultaneously fit with the same model and fixed experimental parameters, apart from the line splitting and amplitude ratio, as shown in Figure 4.3. For the DFG line shape measurements we have minimized  $V_0$  by looking at the BEC spectra shown in Figure 4.3. At this setting the DFG line shape is much broader than the effect of the lattice or, equivalently, the Fermi energy  $E_F \gg V_0$ .



**Figure 4.3** – Absorption spectrum of a BEC in a weak optical lattice for various rotation angles of the polarization of the second ODT beam with respect to the first ODT beam from the configuration shown in Figure 4.2(a). The spectra are offset and centered around the midway frequency of the two lines, and the lines are fits of the time-dependent line shape model. The spectroscopy interaction times used in these measurements are (top to bottom): 1.5 s, 2 s, 1.5 s, 4 s, and 6 s and vary as the Rabi frequency also varies with the rotation angles.



**Figure 4.4** – Double-peak structure of the Bose-Einstein condensate absorption spectrum due to the weak optical lattice measured for a spectroscopy laser probe time of 1 s (top, red) and 3 s (bottom, black). The uncertainty per data point is indicated by the bottom left inset. The full lines are fits of the time-dependent line shape model. For the top (red) and bottom (black) fit we find  $\chi^2 = 0.9$  and  $\chi^2 = 1.3$ , respectively. A single absorption line from the top (red) dataset is used in Figure 4.1.

## 4.5 Determining the $2^{3}S - 2^{1}S$ s-wave scattering length

We measure the time-dependent behavior of the BEC line shapes to extract the scattering length  $a_{ts}$ , which is the only unknown parameter in the line shape calculations. The optical lattice operates with splitting  $\Delta \approx 35$  kHz such that the lattice is as weak ( $V_0 \approx 2E_r^l$ ) as possible but the two lines are separated sufficiently so they can be individually resolved. Background and lifetime measurements provide the one-body loss rate  $\Gamma^{-1} \approx 10$  s and the chemical potential of the BEC at t = 0. The scattering length  $a_{ts}$  is determined by simultaneous fitting of six doublet lines with interaction times ranging from 0.5 s to 3 s (as shown in Appendix 4.D), and Figure 4.4 shows the lines for 1 s and 3 s. The average reduced  $\chi^2$  of all fits is 1.1, showing good agreement of the model with the data. From the fits we find  $a_{ts} = +50(10)_{\text{stat}}(43)_{\text{syst}} a_0$ . The statistical uncertainty is a 1 $\sigma$  uncertainty based on simultaneous  $\chi^2$  minimization of all data sets. The systematic uncertainty is a worst-case error bound based on our estimation of the Rabi frequency  $\Omega_R = 2\pi \times 21(5)$  Hz (see Section 4.B). Our result is in agreement with the estimated range of possible scattering lengths based on previous mean field shift measurements [31]. Furthermore the determination is in agreement with a surprisingly accurate theoretical value  $a_{ts} = +42.5^{+0.5}_{-2.5}a_0$  [269], based on *ab initio* 1  ${}^{3}\Sigma_{g}^{+}$ and 2  ${}^{3}\Sigma_{g}^{+}$  molecular potentials [139] including large ionization widths which make the calculations insensitive to the actual coupling between the potentials. This is discussed in more detail in Appendix 4.D.

#### 4.6 Conclusion

To conclude, we have directly compared the fundamental difference between quantum degenerate fermions and bosons by measuring and calculating the asymmetric absorption line shapes of a Bose-Einstein condensate and a degenerate Fermi gas of metastable helium. The line shape of the Fermi gas shows excellent agreement without any adaptations to the existing model [263]. We extended the line shape of the Bose-Einstein condensate from the existing model [258] to include ac Stark shift and time-dependent depletion of the condensate. The model shows good agreement with the data, and the 2  ${}^{3}S - 2 {}^{1}S$  s-wave scattering length is extracted to be  $a_{ts} = +50(10)_{\text{stat}}(43)_{\text{syst}} a_{0}$ , in good agreement with scattering length calculations.

We also show how a weak optical lattice can induce a line splitting if the lattice recoil is degenerate with the spectroscopy photon recoil. The effect is similar to Bragg scattering and allows observation of the lattice in the optically excited state. Measurement of the line splitting and the total ac Stark shift on the transition frequency would allow determination of both the dynamic polarizability of the ground and excited states. Furthermore, if unresolved, this effect could lead to a frequency broadening or shift in any spectroscopy measurement in an optical dipole trap.

This concludes the first part of this chapter concerning the work as published in [157]. The remainder of this chapter will be devoted to a detailed discussion of several aspects of the experiment in the appendices.

#### 4.A Appendix: BEC line shape model

Using the line shape model as calculated by Killian [258] as a starting point, the ac Stark shift and the time-dependent behavior are introduced in two steps. Finally the analytic solution of the line shape, convolved with a Lorentzian laser line shape, is given.

#### Adding an ac Stark shift

Although the hydrogen experiment is done in a magnetic trap [264, 265], this does not influence the mathematics for the optical dipole trap (ODT) case. The trapping potential of an ODT is proportional to the dynamic polarizability  $\alpha$  and the local intensity  $I(\vec{r})$  of the optical field as  $V(\vec{r}) \propto -\alpha I(\vec{r})$ . As  $I(\vec{r})$  is identical for both atomic states, we can express the potential of the excited state simply as  $V_2(\vec{r}) = (\alpha_2/\alpha_1)V_1(\vec{r})$ , where  $\alpha_1$  and  $\alpha_2$  are the dynamic polarizabilities of the ground and excited states, respectively. In the main paper the ground and excited states are labeled t and s respectively to distinguish between the singlet and triplet states.

The resonance condition of Killian includes a spatially dependent mean field shift and we add another spatially dependent term here. Using the general resonance condition

$$h(\nu - \nu_0) = V_2^{\text{eff}}(\vec{r}) - V_1^{\text{eff}}(\vec{r}), \qquad (4.3)$$

where  $V_{1,2}^{\text{eff}}(\vec{r})$  are the effective potentials of the ground and excited state. Note that the definition of  $\nu$  is slightly different from the definition in Eq. 4.2 in Section 4.3 as we have omitted the ac Stark, Zeeman and recoil shift from the definition for simplicity of notation. Filling in the mean field interaction and the ac Stark shift, we get

$$h(\nu - \nu_0) = \left(\frac{\alpha_2}{\alpha_1} - 1\right) V_1(\vec{r}) + \Delta U n_1(\vec{r}).$$
(4.4)

Here  $\Delta U n_1(\vec{r}) = (U_2 - U_1) n_1(\vec{r}) = (4\pi\hbar^2 n_1(\vec{r}))/m)(a_{21} - a_{11})$  is the mean field interaction term, with atomic mass m, and scattering lengths

 $a_{21}$  and  $a_{11}$  for the excited-ground state and ground-ground state collisions, respectively. The ground state density profile is given by  $n_1(\vec{r})$ . We see how inclusion of the ac Stark effect leads to a spatial dependence of the resonance condition similar to the mean field interactions.

Mean field interactions between two 2  ${}^{1}S_{0}$  atoms are neglected as the estimated fraction of 2  ${}^{1}S_{0}$  atoms is limited to a worst-case upper bound of 0.2% in the measurement. This estimate is based on the fact that the 2  ${}^{1}S_{0}$  atoms experience an antitrapping potential and are therefore expelled from the trapping region at a timescale shorter than 2 ms. We note that this timescale is an upper limit based on worst-case estimates of atoms leaving the trap from the center along the long axial direction. In most cases the timescale is much shorter and on the order to 10  $\mu$ s, which makes the excited state fraction even lower. As the radiative lifetime of the 2  ${}^{1}S_{0}$  state is 20 ms, spontaneous decay does not play a role in this process.

We can use the linear relationship between the ground state trapping potential and the density distribution of the BEC through the Thomas-Fermi relation  $\mu - U_0 = U_1 n_1(\vec{r}) + V_1(\vec{r})$ , where  $U_0 = V_1(r = 0)$  is the depth of the trap. This definition is slightly unconventional, and is caused by the fact that most traps are defined as V(r = 0) = 0 for convenience. In our case we have defined  $V_1(r \to \infty) = 0$ , which leads to the slight modification. Substituting this in Eq. 4.4, we find

$$h(\nu - \nu_0) = \left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) + \left(U_2 - \frac{\alpha_2}{\alpha_1}U_1\right)n_1(\vec{r}).$$
(4.5)

This resonance condition now only depends on the spatial dependence of the density distribution of the BEC, and any further analysis is completely analogous to the work done by Killian. An important check is to see what happens if the trapping potentials are equal, which is the case for the hydrogen experiment or, indeed, a magic wavelength ODT. In this case  $\alpha_1 = \alpha_2$  and Eq. 4.5 indeed reduces to the resonance condition as derived by Killian.

The behavior of the resonance condition can also be visualized in a schematic way as shown in Figure 4.5. Outside of the optical dipole trap the resonance frequency is the unperturbed frequency  $\nu_0$ , but this frequency is ac Stark-shifted in the optical dipole trap due to the differential polarizability of the states (as the polarizabilities  $\alpha_2/\alpha_1 \approx -1.64$ ).



**Figure 4.5** – Schematic overview of the excitation in an energy-position graph. The unperturbed transition frequency  $\nu_0$  is ac Stark shifted due to the different ac polarizabilities  $\alpha_{1,2}$  of the 2  ${}^3S_1$  and 2  ${}^1S_0$  states. In the Thomas-Fermi limit the Bose-Einstein condensate with chemical potential  $\mu$  has a density profile  $n_{BEC}(r,t)$  mimicking the trapping potential with depth  $U_0$ . Due to the spatial dependence of the ac Stark shift and the mean field shift, the atoms are resonant at a range of positions between the frequencies  $\nu_{min}$  and  $\nu_{max}$ , which are explained in the main text. During the excitation atoms leave the Bose-Einstein condensate and lower the chemical potential. As a result also the range of frequencies over which the atoms are resonant becomes narrower and shifts to higher frequencies.

Including the mean field interactions two extremes in the shifted transition frequencies are identified. The center frequency  $\nu_{\text{max}}$  is given as the sum of the maximum ac Stark shift and the maximum mean field shift  $h\nu_{\text{max}} = [(\alpha_2/\alpha_1) - 1](\mu - U_0) + (U_2/U_1 - \alpha_2/\alpha_1)\mu$ , where we use the fact that  $\mu \propto a_{11}n_1(r=0)$ . Similarly, at the edge of the Bose-Einstein condensate (at the Thomas-Fermi radius  $R = \sqrt{2\mu/m\omega^2}$  where  $U_{\text{dip}} = \mu$ ), the density is zero and the only contribution is the ac Stark shift to the minimum shifted transition frequency  $h\nu_{\text{min}} = [(\alpha_2/\alpha_1) - 1](\mu - U_0)$ . As the atoms are excited from the Bose-Einstein condensate for t > 0the condensate shrinks and the chemical potential drops, resulting in a narrower range of blue-shifted (higher) frequencies for which the atoms are resonant.

#### New line shape model

Using the definition by Killian to calculate the line shape for a Dopplersensitive profile in a spherically symmetric trap, we start with

$$S(\nu) = \pi \hbar \Omega_R^2 \int 4\pi r^2 n_1(r) dr \times \delta \Big[ h(\nu - \nu_0) + \Big( \frac{\alpha_2}{\alpha_1} - 1 \Big) (U_0 - \mu) - \Big( U_2 - \frac{\alpha_2}{\alpha_1} U_1 \Big) n_1(r) \Big], \quad (4.6)$$

where  $\Omega_R$  is the Rabi frequency. This integral has the analytical solution

$$S(\nu) = \frac{15\pi\hbar\Omega_R^2 N}{4} \frac{h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu)}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)^2 \mu^2} \times \sqrt{1 - \frac{h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu)}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu}}, \quad (4.7)$$

with N the total number of atoms in the condensate. Eq. 4.7 is also given in Section 4.3 as Eq. 4.2. This function is valid in the frequency domain

$$\left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) \le h(\nu - \nu_0) \le \left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) + \left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu.$$
(4.8)

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This looks quite similar to Eq. 25 from Killian's work [258], which becomes more apparent if the ac Stark effect is removed by setting  $\alpha_1 = \alpha_2$ i.e. the 'magic wavelength' condition. Also note that  $U_2/U_1 = a_{21}/a_{11}$ and therefore this result has the nice feature of being dependent on the - dimensionless - ratios of the polarizabilities and scattering lengths.

#### Depletion of the condensate during excitation

The weak excitation approximation - as used in [258] - assumes that the chemical potential of the condensate does not change significantly during the excitation. In the hydrogen BEC work this is a good approximation as only a fraction of  $10^{-2}$  of the condensate is excited. In our case the excited fraction reaches over 50% and the change of the chemical potential during the excitation has to be taken into account in the line shape calculations.

In the Thomas-Fermi limit the relationship between the chemical potential  $\mu$  and the atom number N of a BEC is the well-known nonlinear relationship

$$N = \frac{2^{\frac{5}{2}}}{15\sqrt{m}\hbar^2 a\bar{\omega}^3} \mu^{\frac{5}{2}},\tag{4.9}$$

where m is the atomic mass, a the s-wave scattering length and  $\bar{\omega} = (\omega_x \omega_y \omega_z)^{\frac{1}{3}}$  the geometric averaged trap frequency. The atom number loss and chemical potential loss are then related as

$$\frac{dN}{dt} = \frac{dN}{d\mu}\frac{d\mu}{dt} = \frac{5}{2}\frac{2^{\frac{3}{2}}}{15\sqrt{m}\hbar^2 a\bar{\omega}^3}\mu^{\frac{3}{2}}\frac{d\mu}{dt} = \frac{5}{2}\frac{N}{\mu}\frac{d\mu}{dt}.$$
 (4.10)

Realizing that the function  $S(\nu)$  represents the one body atom number loss dN/dt, we can use this relationship to use Eq. 4.7 as a differential equation for the chemical potential:

$$\frac{d\mu}{dt} = \frac{2}{5} \frac{\mu}{N} S(\nu) = \frac{3\pi\hbar\Omega_R^2}{2} \frac{h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu(t))}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)^2 \mu} \times \sqrt{1 - \frac{h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu(t))}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu(t)}}.$$
(4.11)

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Solving this differential equation will result in a function  $\mu(\nu, t)$  which shows how the chemical potential of the condensate changes as function of the laser frequency and interaction time.

The above derivation assumes that the condensate stays in equilibrium throughout the excitation, although we can achieve significant excitation fractions in the experiment. To see if the condensate can redistribute fast enough to assume equilibrium, we can compare the change in density distribution to the sound velocity of the condensate. As the density distribution scales linearly with the chemical potential, the estimated upper limit on the relative change in the density distribution is

$$\frac{1}{\mu}\frac{d\mu}{dt} = \frac{2}{5}\frac{1}{N}\frac{dN}{dt} \approx 0.13 \text{ s}^{-1},$$
(4.12)

where we assume a (large) atom number of  $N = 10^6$  atoms and a (short) excitation time of 3 s based on the achieved depletion in Figure 4.4 in Section 4.5. The sound velocity is [202]

$$c = \sqrt{\frac{\mu}{m}} \approx 2 \text{ cm/s}, \qquad (4.13)$$

which, for a condensate with a length of  $\sim 200 \ \mu m$  (which is an upper limit), corresponds to a frequency of the density oscillations of  $100 \ s^{-1}$ . As this is three orders of magnitude larger than the inverse timescale at which the density of the condensate changes, we can assume that the BEC is in equilibrium throughout the excitation.

As the interaction time is on the order of seconds, the condensate also decreases due to one-body collisions with the background gas. This is typically characterized by a loss rate  $\Gamma$ , and the simple differential equation

$$\frac{dN}{dt} = -\Gamma N \tag{4.14}$$

actually leads to a second term in the differential for the chemical potential as

$$\frac{d\mu}{dt} = -\frac{2}{5}\Gamma\mu. \tag{4.15}$$

Two- and three-body losses can, in principle, be implemented in a similar fashion.

#### Adding homogeneous broadening mechanisms

We are not in the limit where the spectral linewidth of the system (~ few kHz) is negligible compared to the observed width of the spectral feature (~ 15 kHz). In order to properly include a homogeneous broadening mechanism, we convolve the initial line shape with a broadening function  $g(\nu)$  as

$$\tilde{S}(\nu) = \int_{\nu'} d\nu' g(\nu - \nu') S(\nu').$$
(4.16)

This means that we do not have to change any of the first principle considerations, but can simply convolve our analytical result of the line shape with any broadening mechanism (which has to be independent of the spatial distribution of the atoms) without changing the formalism.

#### Analytical result for $\tilde{S}(\nu)$

Defining the Lorentzian distribution as

$$g(\nu) = \frac{1}{2\pi} \frac{\gamma}{\nu^2 + (\gamma/2)^2},\tag{4.17}$$

with  $\gamma$  the full-width-half-max (FWHM), we obtain an analytical solution of the convolution with the line shape as defined in Eq. 4.7. We introduce (rescaled) variables to simplify the final expression:

$$\tilde{\mu} \equiv \left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu,\tag{4.18}$$

$$h\tilde{\nu} \equiv h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu),$$
 (4.19)

$$h\Delta \equiv \sqrt{(h\tilde{\nu} - \tilde{\mu})^2 + (h\gamma/2)^2},\tag{4.20}$$

$$h\delta_{\pm} \equiv h\Delta \pm (h\tilde{\nu} - \tilde{\mu}), \qquad (4.21)$$

$$h\xi_{\pm} \equiv \sqrt{(h\gamma)^2 \pm 8h\delta_{\pm}(h\tilde{\nu} - \tilde{\mu})}.$$
(4.22)

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The line shape then becomes

$$\tilde{S}(\nu) = \frac{30}{32} \frac{\hbar \Omega_R^2 N}{\tilde{\mu}^{\frac{5}{2}}} \left[ 2h\gamma \sqrt{\tilde{\mu}} + \left( h\tilde{\nu}\sqrt{h\xi_-} + \frac{h\gamma}{2}\sqrt{h\xi_+} \right) \right] \\ \times \left( \tan^{-1} \left[ \frac{-\sqrt{2\delta_+\tilde{\mu}/h}}{2\Delta - \sqrt{2\delta_-\tilde{\mu}/h}} \right] - \tan^{-1} \left[ \frac{\sqrt{2\delta_+\tilde{\mu}/h}}{2\Delta + \sqrt{2\delta_-\tilde{\mu}/h}} \right] \right) \\ - \left( h\tilde{\nu}\sqrt{h\xi_+} - \frac{h\gamma}{2}\sqrt{h\xi_-} \right) \left( \log \left[ \sqrt{\frac{h\delta_+\tilde{\mu}}{2(h\Delta)^2} + \left( 1 - \sqrt{\frac{h\delta_-\tilde{\mu}}{2(h\Delta)^2}} \right)} \right] \right] \\ - \log \left[ \sqrt{\frac{h\delta_+\tilde{\mu}}{2(h\Delta)^2} + \left( 1 + \sqrt{\frac{h\delta_-\tilde{\mu}}{2(h\Delta)^2}} \right)} \right] \right) \right]. \quad (4.23)$$

This expression is used in Eq. 4.11 to calculate the time-dependent line shape used to fit the data as shown in Figures 4.1, 4.3, 4.4, and 4.14. It is interesting to note that most of the parameters in the full analytical expression 4.23 are fixed or can be determined in our experiment with sufficient accuracy. For determining the 2  ${}^{3}S_{1} - 2 {}^{1}S_{0}$  scattering length the limiting factor in the accuracy turns out to be the Rabi frequency  $\Omega_{R}$ , as the overall prefactor of the lineshape contains the ratio  $\Omega_{R}^{2}/\tilde{\mu}^{5/2}$ and using Eq. 4.18, there is a near-linear dependence between the scattering length  $a_{21}$  and  $\Omega_{R}$ . The accuracy in determining  $a_{21}$  is therefore limited by the accuracy of our estimate of the Rabi frequency, which will be discussed in Appendix 4.B.

#### Comparing the line shape model results

The model as presented by Killian [258] is now extended to include ac Stark shift and time-dependent effects, and it is interesting to see what sort of line shapes can be expected for the current experimental parameters. It is also interesting to see how the inclusion of additional effects, such as the one-body loss and the laser linewidth, affect the line shapes. Therefore the line shapes for three different cases are shown in Figure 4.6, starting with the most basic model, Figure 4.6 (a) and (b), with added one-body losses in Figure 4.6 (c) and (d), and finally the full model including the spectroscopy laser linewidth in Figure 4.6 (e) and (f).

For short interaction times the line shape looks similar to the solutions developed by Killian [258], but the most striking effect is the formation of a sawtooth-like line shape for longer interaction times. This is caused by the fact that the condensate shifts more towards the 'blue' side of the spectrum as it is depleted of atoms (i.e. the chemical potential decreases towards the bottom of the trap, as schematically shown in Figure 4.5). Its instantaneous line shape still looks like the weak-interaction line shape, but when integrated over time it creates a sawtooth-like shape.

As a result of this effect, the line shape as observed in the experiment (broadened by the spectroscopy laser linewidth of 4.5 kHz) will show a systematic shift to higher frequencies with increasing interaction time if the time-dependent behavior is not taken into account, as we can see in Figure 4.6 (e) and (f). Furthermore, the asymmetry in the line shape caused by the mean field interactions is reduced significantly, which is why the line shape is measured at multiple interaction times to determine the scattering length.

#### 4.B Appendix: Estimating the Rabi frequency

As mentioned in the previous section, the accuracy of the determined  $2 {}^{3}S_{1} - 2 {}^{1}S_{0}$  scattering length is limited by the accuracy with which the Rabi frequency of the optical transition is determined. Therefore the exact quantization axis (i.e. the direction of the magnetic field) of the atom is calculated together with the projection of the spectroscopy beam along (and orthogonal to) this axis in order to calculate the Rabi frequency accurately enough for the scattering length determination. First the coordinate system and the magnetic fields are defined, after which the geometric parameters (i.e. polar and azimuthal angles of the magnetic field) can be calculated. The projection of the spectroscopy beam onto the magnetic field can be calculated next in order to estimate the Rabi frequency. Finally the effect of the unknown ellipticity of the polarization (due to birefringence of the vacuum windows) on the Rabi frequency is estimated.

4.



Figure 4.6Time-dependent behavior of the line shape of a Bose-Einstein condensate using typical experimental parameters:  $\Omega_R = 40\pi \ rad/s, \ \Gamma = \Omega_R/200, \ \alpha_2/\alpha_1 = -1.64, \ and \ a_{ts}/a_{ss} = 50/142.$ The initial chemical potential of the condensate is  $\mu/k_B = 0.2 \ \mu K$ . Each figure shows the line shape for interaction times of  $\Omega_R t = 20\pi, 40\pi, 60\pi$ , and  $80\pi$  normalized to the background level at the shortest interaction time shown. The figures on the left-hand side show the line shape as function of the chemical potential  $\mu(\nu)$ , and on the right-hand side as function of the atom number  $N(\nu)$  of the condensate. Figures (a) and (b) show the line shape by solving Eq. 4.11. Figures (c) and (d) show the line shape by solving Eq. 4.11 and including one-body loss as defined by Eq. 4.15. Figures (e) and (f) show the line shapes when including a spectroscopy laser linewidth of 4.5 kHz following Eq. 4.23 and one-body loss. For short interaction times the line shapes look very similar to the analytical solution given by Killian [258], but evolve into a distinct sawtooth shape. This effect is smeared out significantly with the (current) spectroscopy laser linewidth. 126

#### Defining the geometry and calculating the field

The basic coordinate system is defined as shown in Figure 4.7, where the z-axis is aligned with the MOT z-beam in our experiment, and therefore represents the axial direction (longitudinal axis) of the ODT and the symmetry axis of the fine tune field. As the quantization axis is given by the magnetic field, the magnetic field in this coordinate system is calculated and used to project the spectroscopy beam on in a later stage. The background magnetic field

$$\vec{B}_{bg} = B^x_{bg}\vec{e}_x + B^y_{bg}\vec{e}_y + B^z_{bg}\vec{e}_z$$
(4.24)

is defined as

$$B_{bg}^{x} = B_{0} \sin(\theta) \cos(\phi),$$
  

$$B_{bg}^{y} = B_{0} \sin(\theta) \sin(\phi),$$
  

$$B_{bg}^{z} = B_{0} \cos(\theta).$$
  
(4.25)

Here  $B_0 = \nu_0/\gamma$  is the magnitude of the magnetic field which can be measured using rf spectroscopy where  $\nu_0$  is the resonance frequency (in Hz) and  $\gamma = 2.8$  MHz/G the magnetic moment of the state. Applying a magnetic field along the z-axis

$$\vec{B}_{ext} = B_{ext}\vec{e}_z,\tag{4.26}$$

the new magnetic field  $\vec{B}_{new}$  is

$$B_{new}^{x} = B_{0} \sin(\theta) \cos(\phi),$$
  

$$B_{new}^{y} = B_{0} \sin(\theta) \sin(\phi),$$
  

$$B_{new}^{z} = B_{0} \cos(\theta) + B_{ext}.$$
  
(4.27)

This is rewritten in a form with a new magnitude  $B'_0$  and polar angle  $\theta'$  as

$$B_{new}^{x} = B'_{0} \sin(\theta') \cos(\phi),$$
  

$$B_{new}^{y} = B'_{0} \sin(\theta') \sin(\phi),$$
  

$$B_{new}^{z} = B'_{0} \cos(\theta'),$$
  
(4.28)

by realizing that

$$B_0 \cos(\theta) + B_{ext} = B'_0 \cos(\theta'), \qquad (4.29)$$

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**Figure 4.7** – Schematic overview of our coordinate system, where the z-direction is aligned with the axial direction (longitudinal axis) of the ODT. The polar angle is  $\theta$  and azimuthal angle is  $\phi$ . The radial length r is equivalent to the magnitude of the field e.g.  $r = |\vec{B}_0|$ .

and that the magnitude of the new field is

$$B_0^{'2} = (B_0 \cos(\theta) + B_{ext})^2 + B_0^2 \sin^2(\theta),$$
  

$$B_0^{'2} = B_0^{'2} \cos^2(\theta') + B_0^2 \sin^2(\theta).$$
(4.30)

This defines the new angles

$$\cos^{2}(\theta') = 1 - \left(\frac{B_{0}}{B'_{0}}\right)^{2} \sin^{2}(\theta),$$
  

$$\sin^{2}(\theta') = \left(\frac{B_{0}}{B'_{0}}\right)^{2} \sin^{2}(\theta).$$
(4.31)

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Thus the new magnetic field can be described as

$$B_{new}^{x} = B_{0}' \cos(\phi) \sqrt{\left(\frac{B_{0}}{B_{0}'}\right)^{2} \sin^{2}(\theta)},$$
  

$$B_{new}^{y} = B_{0}' \sin(\phi) \sqrt{\left(\frac{B_{0}}{B_{0}'}\right)^{2} \sin^{2}(\theta)},$$
  

$$B_{new}^{z} = B_{0}' \sqrt{1 - \left(\frac{B_{0}}{B_{0}'}\right)^{2} \sin^{2}(\theta)}.$$
(4.32)

As both  $B_0$  and  $B'_0$  can be measured using rf spectroscopy (using  $B_0/B'_0 = \nu_0/\nu'_0$ ), the new angle  $\theta'$  can be determined directly if the original angle  $\theta$  is known. No information about the azimuthal angle  $\phi$  is obtained as the magnetic field is only applied along the z-axis and thereby not breaking the symmetry.

#### Measuring the polar angle $\theta$ for $B_{bq}$

The derived magnetic field relations can be used to measure  $\theta$  by applying an external magnetic field  $B_{ext}$  and observing how the total magnetic field amplitude  $B'_0$  (i.e. the rf resonance frequency) changes as function of this field. Knowing that

$$|B_0'| = \sqrt{(B_0 \cos(\theta) + B_{ext})^2 + B_0^2 \sin^2(\theta)},$$
  

$$|B_0'| = \sqrt{B_0^2 + B_{ext}^2 + 2B_0 B_{ext} \cos(\theta)},$$
(4.33)

the fine tune field calibration measurements can be used for a simple fit of the latter equation to determine the angle

$$\cos(\theta) = 0.614(6),$$
  
(4.34)  
$$\theta = 52.1(4)^{\circ}.$$

#### 'Flipping' the quantization axis

By applying an external magnetic field pointing into the  $-\vec{e}_z$  direction (i.e.  $\vec{B}_{ext} = -B_{ext}\vec{e}_z$ ), the new total magnetic field gives a resonance

frequency  $\nu'_0 \approx 1.682$  MHz (without external field it is  $\nu_0 = 1.4635$  MHz). In this case the new angle  $\theta'$  is

$$\sin(\theta') = \Big| -\frac{1.4635}{1.682} \sin(\theta) \Big|, \qquad (4.35)$$
  
$$\theta' = 136.6(3)^{\circ}.$$

This means that the new magnetic field vector is pointing in the opposite direction along the z-axis i.e. the quantization axis has flipped in the z-direction (but not along the azimuthal coordinate  $\phi$ ). This can be used as a method to check for systematic effects depending on the absolute orientation of the quantization axis.

#### From magnetic field to quantization axis

The quantization axis generally is a unit vector defining the orientation of the field. In our case the quantization axis is defined by the magnetic field, and in order to talk about a clean quantization axis, the vectors as described in the previous sections should be used, but they are divided by the magnitude of the magnetic field such that  $\vec{Q} = \vec{B}/|\vec{B}|$ . This is a unit vector which is cleaner to deal with when considering the projection of the spectroscopy light onto the quantization axis.

#### Defining the spectroscopy light

The spectroscopy light is defined by a k-vector  $\vec{k}$  and the polarization of the light (i.e. the orientation of the electric field  $\vec{E}$ ), which is orthogonal to  $\vec{k}$ . Although the the electric field is considered in this example, this approach also works for the magnetic field component (which is orthogonal to both  $\vec{k}$  and  $\vec{E}$ ) which is relevant for magnetic dipole transitions. The spectroscopy beam is defined as  $\vec{k} = k_y \vec{e}_y + k_z \vec{e}_z = (2\pi/\lambda) \times$  $(\sin(\alpha)\vec{e}_y + \cos(\alpha)\vec{e}_z)$  when traveling in the y-z plane with an angle  $\alpha = -9.5^{\circ}$  with respect to the z-axis. The polarization of the electric field  $\vec{E} = E^x \vec{e}_x + E^y \vec{e}_y + E^z \vec{e}_z$  is orthogonal to  $\vec{k}$ . Defining a azimuthal coordinate  $\phi'$  which defines the orientation of the polarization in the (x, y, z)-space with  $\phi' = 0$  in the x-z plane if  $\alpha = 0$  (i.e. for  $\phi' = \pi/2$ the polarization is pointing along the y-axis, similar to the definition of  $\phi$  in Figure 4.7), gives

$$E^{x} = E_{0} \cos(\phi'),$$
  

$$E^{y} = E_{0} \cos(\alpha) \sin(\phi'),$$
  

$$E^{z} = -E_{0} \sin(\alpha) \sin(\phi').$$
  
(4.36)

The angles  $\alpha$  and  $\phi'$  are in principle known, apart from the fact that the birefringence of the windows might change  $\phi'$ .

#### Projecting the spectroscopy light

The goal of all these calculations is to figure out how much of the spectroscopy light is actually used for making a transition. In the experiment a  $\sigma^-$  transition is induced, which actually depends on the orthogonal part of the field with respect to the quantization axis. The projection *along* the quantization axis is calculated, from which the remaining (i.e. orthogonal) part can be deduced.

The normalized projection P of vector  $\vec{E}$  on quantization axis vector  $\vec{Q}$  is

$$P = \frac{\vec{E} \cdot \vec{Q}}{|\vec{E}|} = \frac{1}{E_0} \sum_{i=x,y,z} E^i Q^i = \frac{1}{E_0 B_0} \sum_{i=x,y,z} E^i B^i,$$
(4.37)

which is

$$P = \sin(\theta)\cos(\phi)\cos(\phi') + \sin(\theta)\sin(\phi)\cos(\alpha)\sin(\phi') -\cos(\theta)\sin(\alpha)\sin(\phi'). \quad (4.38)$$

In this expression only  $\phi$  is completely unknown and the other angles can be measured or fully controlled. The projection P is along the quantization axis and is relevant for  $\pi$  transitions. For the  $\sigma^-$  transitions the orthogonal projection  $P' = \sqrt{1 - P^2}$  is the relevant parameter. The Rabi frequencies  $\Omega_R$  scale linearly with this projection but the magnitude (i.e. depletion) of the line shapes scale with  $\Omega_R^2$  and therefore with  $P'^2$  (or, equivalently,  $1 - P^2$ ).



Figure 4.8 – Orthonormal projection  $P'^2$  for the unknown quantization axis azimuthal angle  $\phi$ , assuming the quantization axis polar angle  $\theta = 52.1^{\circ}$  and the spectroscopy light polarization rotation angle  $\phi' = -16^{\circ}$  as used in the experiments.

#### Application to our measurements

Figure 4.8 shows the variation in  $P'^2$  for the full range of possible values of the quantization axis azimuthal angle  $\phi$ , given an experimentally determined polar angle  $\theta = 52.1^{\circ}$  and a spectroscopy light polarization rotation angle of  $\phi' = -16^{\circ}$  as used in the experiment.

 $P'^2$  - and thereby  $\Omega_R^2$  - varies from 0.4 to 1.0. As there is no additional information to reduce this range, a systematic uncertainty is set on the projection as  $P'^2 = 0.7 \pm 0.3$  (i.e.  $\sim 43\%$  uncertainty). It should be noted that this uncertainty covers the full range of possible values i.e. it is a conservative estimate of the uncertainty.

This calculated range of effective projections provides an estimate of the Rabi frequency  $\Omega_R$  which is defined as

$$\Omega_R^2 = \frac{6\pi c^2}{\hbar\omega_0^3} A_{21} |\langle J_1 M_1 | q | J_2 M_2 \rangle|^2 I_0.$$
(4.39)

Here  $\omega_0$  is the transition frequency,  $A_{21} = 9.1 \times 10^{-8} \text{ s}^{-1}$  the Einstein coefficient as calculated by Pachucki *et al.* [106],  $I_0 = 2P_0/\pi w_0^2$  the peak intensity with  $P_0$  the total power and  $w_0 = 0.3$  mm the beam waist of the spectroscopy beam. The remaining matrix element  $|\langle J_1 M_1 | q | J_2 M_2 \rangle|^2 = C_{C-G}^2 \cdot P'^2$ , where q = -1, 0, +1 represents the possible transitions (i.e.  $\sigma^-, \pi, \sigma^+$ , respectively),  $C_{C-G}^2 = 1/3$  the corresponding Clebsch-Gordan coefficient of the transitions, and  $P'^2$  the relevant projection of the spectroscopy light onto the quantization axis. As the  $m = +1 \rightarrow m' = 0$  transition is currently under consideration, q = -1 and  $P'^2$  represents the projection orthogonal to the quantization. An additional factor  $(1/\sqrt{2})^2$  is added to this term, as the orthogonal projection H of the linearly polarized light is equally decomposed in left-handed (LCP) and right-handed (RCP) circular polarized light as  $|H\rangle = (|LCP\rangle + |RCP\rangle)/\sqrt{2}$ .

All these elements are combined to calculate the Rabi frequency. Just for reference the calculation is split for the different terms:

$$\frac{6\pi c^2}{\hbar \omega_0^3} A_{21} = 0.28 \text{ m}^2/\text{J s}, \qquad (4.40)$$

and

$$I_0 = \frac{2P_0}{\pi w_0^2} = \frac{2 \cdot P'_0 \cdot T \cdot \chi}{\pi w_0^2} = 5.5 \times 10^5 \text{ W/m}^2, \qquad (4.41)$$

using a typical measured power of  $P'_0 \approx 95$  mW, T = 0.9 is the transmission of the vacuum window,  $\chi = 0.9$  represents additional reflection losses between the position where the optical power is measured and the vacuum window. Including the Clebsch-Gordan coefficients and the factor 1/2, the Rabi frequency is  $\Omega_R^2 = 5.1 \times 10^4 \times \frac{P'^2}{2}$  (rad/s)<sup>2</sup>. Using the calculated range for  $P'^2 = 0.7 \pm 0.3$  gives

$$\Omega_R = (1.33 \pm 0.29) \times 10^2 \text{ rad/s} = 2\pi \times (21 \pm 5) \text{ Hz.}$$
(4.42)

#### Checking the polarization of our spectroscopy light

The observations of the apparent presence of an optical lattice in the ODT means that the polarization of the laser beams is slightly rotated (and possibly made elliptical) due to birefringence of the vacuum windows. It is important to know how much the linear polarization changes, as it affects the effective Rabi frequency. This effect can be quantified by considering the following.

Any arbitrary polarization state  $\overline{M}$  can be described in a x - y basis as

$$\bar{M} = \begin{bmatrix} E_x \\ E_y \end{bmatrix} = \begin{bmatrix} A \\ B \pm iC \end{bmatrix}, \qquad (4.43)$$

where the  $\pm$  sign represents the handedness of the polarization. This is not known for now, but the sign can be kept as such as it will drop out in the later calculations. This polarization state can be decomposed into normalized linear and circular polarization components as

$$\begin{bmatrix} A\\ B\pm iC \end{bmatrix} = c_0 \begin{bmatrix} 1\\ 0 \end{bmatrix} + c_1 \begin{bmatrix} 0\\ 1 \end{bmatrix} + \frac{c_2}{\sqrt{2}} \begin{bmatrix} 1\\ \pm i \end{bmatrix}, \qquad (4.44)$$

resulting in the following relations

$$c_0 = A - C,$$
 (4.45)

$$c_1 = B, \tag{4.46}$$

$$c_2 = \sqrt{2C}.\tag{4.47}$$

The arbitrary polarization state can be written as

$$\begin{bmatrix} A \\ B \pm iC \end{bmatrix} = (A - C) \begin{bmatrix} 1 \\ 0 \end{bmatrix} + B \begin{bmatrix} 0 \\ 1 \end{bmatrix} + C \begin{bmatrix} 1 \\ \pm i \end{bmatrix}.$$
 (4.48)

Furthermore the polarization state is required to be normalized, i.e.  $|E_x|^2 + |E_y|^2 = A^2 + B^2 + C^2 = 1$ . Explicit calculation of  $\overline{M}^{\dagger}\overline{M}$  using the decomposition as given in Eq. 4.48 shows that this is indeed the case.

Rotating this polarization state using a half-wave plate, the final polarization state is given by

$$\begin{bmatrix} E'_x \\ E'_y \end{bmatrix} = \begin{bmatrix} \cos(\phi') & -\sin(\phi') \\ \sin(\phi') & \cos(\phi') \end{bmatrix} \begin{bmatrix} A \\ B \pm iC \end{bmatrix}, \quad (4.49)$$

where  $\phi'/2$  is the angle of rotation of the half-wave plate (following the definition of  $\phi'$  as the polar angle of the spectroscopy polarization).



**Figure 4.9** – Normalized transmission of a polarizing beam splitter before (left) and after (right) the vacuum chamber whilst rotating a halfwave plate. The solid lines are fits using Eq. 4.51.

**Table 4.1** – Polarization state parameters determined by fitting Eq. 4.51 to the data as shown in Figure 4.9. The polarization state has a slight deviation in the determined normalization condition (bottom right), which is explained in the main text.

	Vacuum chamber	
Parameter	Before	After
R	42(4)	4.2(1)
$A^2$ (from $R$ )	0.977(2)	0.808(4)
$B^2 + C^2$ (from $R$ )	0.023(2)	0.192(4)
B (fit)	$\sim 0$	0.16(3)
C (fit)	0.150(7)	0.454(6)
$C^2$ (fit)	0.023(2)	0.207(5)
$B^2 + C^2$ (fit)	0.023(2)	0.23(1)
$A^2 + B^2 + C^2$ (fit)	0.999(3)	1.04(1)

The new x-component is

$$E'_{x} = A\cos(\phi') - (B \pm iC)\sin(\phi'), \qquad (4.50)$$

or, considering the intensity in stead of the field

$$|E'_{x}|^{2} = A^{2}\cos(\phi')^{2} - 2AB\cos(\phi')\sin(\phi') + (B^{2} + C^{2})\sin(\phi')^{2}.$$
 (4.51)

It is this behavior that is expect to be observed if the transmission through a polarizing beam splitter is measured when rotating an elliptic polarization state. Figure 4.9 shows the normalized transmission of the optical dipole trap beam as measured using a polarizing beam splitter before and after the vacuum chamber. The solid lines are fits to the data using Eq. 4.51.

It is clear that the polarization is no longer linear after the vacuum chamber, as visibility of the fringes has decreased significantly. From these measurements we can already determine most of the parameters A, B, and C of the polarization state.

The ratio  $R = |E_x|^2/|E_y|^2$ , which represents the ratio of the major and minor axis of the elliptic polarization, is determined by the ratio of the maximum and minimum transmission of the light and is given in Table 4.1. From the matrix representation this ratio can be written as

$$R = \frac{|E_x|^2}{|E_y|^2} = \frac{A^2}{B^2 + C^2}.$$
(4.52)

Combined with the normalization condition this gives

$$A^2 = \frac{1}{1+1/R},\tag{4.53}$$

$$B^2 + C^2 = \frac{1}{1+R}. (4.54)$$

The values of R are known from the fits (see Table 4.1) the value for A can be filled in and this parameter can be fixed in the fits for B and C. Using this method provides values for B and C from the fits, and can be cross-checked with the expected value for  $B^2 + C^2$  and the normalization condition. There is good but not excellent agreement as there is a deviation from the experimentally determined normalization of the polarization state after transmission throught the vacuum chamber (see Table 4.1). This is probably because this approach neglects nonlinear effects that also play a role (such as slight ellipticity introduced in the polarization by non-Brewster reflection off a mirror). However, it is sufficient to provide an estimate of the ellipticity of the polarization.

The parameter  $C^2$  describes the 'amount' of circular polarization in the beam, and this constitutes about 2.3(2)% prior to the vacuum chamber and 20.7(5)% after the vacuum chamber i.e. a nine-fold increase. As the handedness of the circular polarization is not known, this percentage either presents a loss or increase of signal<sup>1</sup>.

This can be parametrized by defining the new  $\sigma^-$ -projection  $\tilde{P}^2$ , which is defined as <sup>2</sup>

$$\tilde{P}^2 = \frac{P'^2}{2}(1 - C^2) + C^2, \qquad (4.55)$$

$$\tilde{P}^2 = \frac{P^2}{2} (1 - C^2), \qquad (4.56)$$

depending whether the circular polarization is allowed to induce a transition or not. In this definition and in the limit of  $C^2 \rightarrow 0$ ,  $\tilde{P}^2$  goes to  $P'^2/2$  (so the factor of 2 needs to be taken into account if the Rabi frequency should be recalculated). The new effective orthogonal projection is shown in Figure 4.10. The range of determined values of  $C^2$  gives  $0.275 \leq \tilde{P}^2 \leq 0.48$  (or,  $0.55 \leq P'^2 \leq 0.96$ ). This is within the earlier calculated range of  $P'^2/2 = (0.7 \pm 0.3)/2 = 0.35 \pm 0.15$  where a pure linear polarization was assumed. Further analysis could be done to narrow this range down, but as it is already smaller than the conservative systematic error bar calculated before it is not essential to pursue this.

<sup>&</sup>lt;sup>1</sup>The Clebsch-Gordan coefficients for all transitions in this case are equal, but we cannot make  $\sigma^+$  transitions from the m = +1 state due to selection rules. So if the handedness allows a  $\sigma^-$  transition, it presents an increase in signal but the opposing situation would present a loss of signal.

<sup>&</sup>lt;sup>2</sup>For  $C^2 \to 0$  the value of  $\tilde{P}^2 \to P'^2/2$ , as only 50% of the orthogonal linear polarization can be used for a  $\sigma^-$  transition. But for  $C^2 \to 1$  either all or none of the orthogonal polarized light can be used for the transition.



**Figure 4.10** – New effective projection  $\tilde{P}^2$  recalculated for the two cases if circular polarization is present (for either handedness). From our analysis we know that  $0.023 \leq C^2 \leq 0.207$  (vertical lines), and within that range we have  $0.275 \leq \tilde{P}^2 \leq 0.48$  (horizontal lines).

#### Concluding remarks about the Rabi frequency

In this Appendix the full three-dimensional orientation of the magnetic field and the spectroscopy beam field are calculated and determined. From magnetic field scans using the fine tune coils the polar angle of the magnetic field could be constrained to  $\theta = 52.1(4)^{\circ}$  with respect to the z-axis in our experiment.

Using the angle of incidence  $\alpha = -9.5^{\circ}$  of the spectroscopy beam with respect to the z-axis and the known polarization angle  $\phi' = -16^{\circ}$ of the spectroscopy light, the orthogonal projection (squared) of the spectroscopy light onto the quantization axis is estimated to be  $P'^2 =$  $0.7 \pm 0.3$ , without using any assumption on the possible azimuthal angle  $\phi$  of the quantization axis. This is the most conservative estimate of this projection. For the typical spectroscopy beam power used in the experiment this gives a Rabi frequency of  $\Omega_R = (1.33 \pm 0.29) \times 10^2$  rad/s =  $2\pi \times (21 \pm 5)$  Hz.
This analysis is based on the assumption that the spectroscopy light is linearly polarized. The effect of possible ellipticity of the polarization due to the birefringence of the vacuum windows is also investigated and provides a conservative estimate of  $0.55 \leq P'^2 \leq 0.96$ . This is within the previously determined bounds (which are also conservative).

#### 4.C Appendix: An optical lattice

To make sure that the two laser beams that constitute our crossed-beam optical dipole trap have orthogonal polarizations, a Glan polarizer and a polarizing beam splitter are used. However, the beams are sent into the vacuum chamber at an angle of incidence of  $9.5^{\circ}$ . As the sapphire windows of our vacuum chamber are birefringent (with the fast axis perpendicular to the window plane), this non-orthogonal incidence creates a slight ellipticity of the polarization in both beams, which are not orthogonal with respect to each other.

A solution would be to use an AOM to detune the 'returning beam' sufficiently so interference does not occur regardless of polarization. This would however require a full reconstruction of the optical dipole trap, which is very time consuming. Furthermore it was noticed before that at the typical ODT powers the AOM would actually cause thermal lensing, making it experimentally difficult to create an ODT with two identical beams.

It is necessary to estimate the typical lattice parameters as used in the experiment to determine in what regime of optical lattice physics the Bose-Einstein condensate and line shape model can be described.

#### Physics in an optical lattice

Typical experimental values of the optical lattice parameters are given in Table 4.2. The discussion is primarily based on an introduction to optical lattices in Refs. [270] (p. 477) and [202] (Ch. 16).

Parameter	Value		
ODT wavelength $\lambda_{\text{ODT}}$	1557.3 nm		
Effective lattice wavenumber $q = 2\pi \cos(\theta/2)/\lambda_{\text{ODT}}$	$3.980 \times 10^6 \text{ m}^{-1}$		
Lattice spacing $d = \pi/q$	789 nm		
Lattice recoil energy $E_r$	20.0 kHz		
Spectroscopy photon recoil energy $E_r^s$	20.6 kHz		
Scattering length $(2 \ {}^{3}S_{1} - 2 \ {}^{3}S_{1}) a_{s}$	7.512(5) nm [152]		
Scattering length compared to lattice spacing $a_s/d$	0.0095		
Chemical potential $\mu$	$0.2 \ \mu \text{K}, \ 4.2 \ \text{kHz}, \ 0.21 \ E_r$		
BEC interaction parameter $g = 4\pi \hbar^2 a_s/m$	$1.56 \times 10^{-49} \text{ J m}^3$		
BEC peak density $n_0 = \mu/g$	$\sim 10^{19} \text{ m}^{-3}$		
Healing length $\xi = \hbar / \sqrt{2mgn_0}$	732 nm		
Healing length compared to lattice spacing $\xi/d$	0.93		
Axial BEC length $l_{\rm ax} = \sqrt{2\mu/m\omega_{\rm ax}^2}$	$\sim 1 \times 10^{-4}~{\rm m}$		
Number of lattice sites $N_{\text{latt}} = l_{\text{ax}}/d$	$\sim 126$		
Lattice harmonic oscillator length $a_{\text{latt}} = \sqrt{\hbar/m\omega_z}$	$\sim 2\times 10^{-7}~{\rm m}$		
$a_{\text{latt}}$ compared to lattice spacing $a_{\text{latt}}/d$	0.25		
$a_{\text{latt}}$ compared to scattering length $a_{\text{latt}}/a_s$	27		
Lattice momentum states $p_l = 2\pi\hbar l/d$	$l\times 8.4\times 10^{-28}~{\rm kg}~{\rm m}~{\rm s}^{-1}$		
BEC atom number $N_0$	$\sim 10^5$		
BEC atoms per site $N_0/N_{\text{latt}}$	$\sim 10^3$		

 Table 4.2 – A list of relevant BEC or optical lattice parameters.

The simple interference pattern created by two counterpropagating beams along the z-axis is defined as

$$V_{\text{latt}}(z) = sE_r \sin^2(qz), \qquad (4.57)$$

where  $E_r = \hbar^2 q^2/2m \approx k_B \times 0.94 \ \mu \text{K} = h \times 20.0 \text{ kHz}$  is the lattice recoil energy,  $q = 2\pi/\lambda_{\text{ODT}}$  and  $s = V_0/E_r$  the dimensionless lattice amplitude. If the two counterpropagating laser beams intersect at an angle  $\theta$ with respect to each other q is redefined as  $q = 2\pi \cos(\theta/2)/\lambda_{\text{ODT}}$ . In a similar way the lattice period  $d = \lambda_{\text{ODT}}/2\cos(\theta/2)$  (and  $q = \pi/d$ ). These are the same definitions as used in Section 4.4.

In such a 1D lattice, the local axial trap frequency per lattice site is  $\hbar\omega_z = h\nu_z = 2\sqrt{s}E_r$  (the radial trap frequency is ~ 250 Hz), and should be compared to the typical chemical potential of the BEC, which is around  $k_B \times 0.2 \ \mu\text{K}$  in our experiment, or 0.21  $E_r$ . Even for a value of s = 1 (which would be a very weak optical lattice), we have  $\mu \ll \hbar\omega_z$ and  $\mu \gg \hbar\omega_{\text{rad}}$ . This shows that the BEC can be described in the lattice using a radial continuum of states, and only using the lowest axial harmonic oscillator level.

#### Band structure

An optical lattice will lead to a band structure which needs to be considered. In the weak interaction regime the band structure of the lowest two bands is given as (Eq. 19 [197])

$$\frac{E(q')}{E_r} = \left(\frac{q'}{q} - 1\right)^2 \pm \sqrt{4\left(\frac{q'}{q} - 1\right)^2 + \frac{s^2}{16}},\tag{4.58}$$

where q' is the quasimomentum within the lattice, and the +(-) sign corresponds to first (lowest) band. As the band structure is periodic over the quasimomentum  $2\pi/d$  (= 2q), only the first Brillouin zone is shown in Figure 4.11. Also shown in Figure 4.11 is the bandgap  $\Delta$ , which is the energy difference between the lowest and first band at the edge of the Brillouin zone (i.e. for  $q = \pi/d = q'$ ), as function on the dimensionless trap depth s. In this regime the bandgap follows the simple proportional relationship  $\Delta/E_r = s/2$ , which is equal to  $\Delta = V_0/2$ .



**Figure 4.11** – Left: band structure in the weak interaction regime calculated from Eq. 19 [197] using s = 2. Right: bandgap  $\Delta$  at  $q = \pi/d$  between the bottom two bands as function of the dimensionless trap depth s.

#### Superfluidity and the Mott insulator regime

There are two main parameters which describe most of the physics occurring in an optical lattice, the hopping parameter J and the on-site interaction parameter U. In the tight-binding approximation  $(s \gg 1)$ and neglecting interaction energies apart from a two-atom on-site interaction energy, they are defined as

$$\frac{J}{E_r} \approx \frac{4}{\sqrt{\pi}} s^{3/4} e^{-2\sqrt{s}},$$
 (4.59)

$$\frac{U}{E_r} \approx \sqrt{\frac{8}{\pi}} a_s q' s^{3/4},\tag{4.60}$$

$$\frac{U}{J} \approx \frac{a_s q'}{\sqrt{2}} e^{2\sqrt{s}},\tag{4.61}$$

where  $a_s$  is the *s*-wave scattering length between the atoms. Figure 4.12 shows the behavior of these parameters for a typical range of lattice depths *s*. These expressions become less valid<sup>3</sup> when  $s \approx 1$  as the (Wannier) wavefunctions become less localized, and analytical results in this regime do not seem to be readily available. In the weak-binding

<sup>&</sup>lt;sup>3</sup>For instance, the hopping parameter J goes to zero in the limit of  $s \to 0$  which does not make sense as the lattice disappears and hopping should be dominant. Of course hopping becomes very ill-defined in this limit.



**Figure 4.12** – Left: lattice parameters  $J/E_r$  (full red line),  $U/E_r$  (dashed blue line) plotted for our lattice and atomic parameters from Table 4.2. The ratio U/J is shown in the figure right. These graphs are only valid for  $s \gg 1$ .

limit ( $s \ll 1$ ) a special regime can be entered if  $\mu \gg sE_r$ , at which the Bloch bands are no longer well defined and a 'swallow-tail' band structure emerges [271]. However, as  $\mu \approx 0.21E_r$  (see Table 4.2) this regime is not accessed.

In the weak-binding regime, lattices with decreasing amplitude have increased hopping J, which promotes delocalization of atoms over the lattice, leading to the superfluid state [272]. In this limit of  $J \gg U$ , the many-body ground state can be considered as an ideal BEC with all atoms in the q' = 0 Bloch state of the lowest band. Technically this state should be described as a coherent superposition of all lattice sites, but this is indistinguishable from the regular BEC state (also see discussion in Sec. IV-B of Ref. [121]).

This limit exists for  $U/J \rightarrow 0$ , and it is interesting to see what happens when this ratio increases as the optical lattice power is increased. In the limit of  $J \ll U$  (tight-binding), hopping becomes less favorable and at some point the Mott insulator state is reached. This transition happens at a critical value of  $(U/J)_c = 5.8z$  for  $\bar{n} = 1$  or  $(U/J)_c = 4z\bar{n}$  for  $\bar{n} \gg 1$ , where  $\bar{n}$  is the average atom number per lattice site, and z the number of nearest neighbours (z = 2 for an optical lattice) [121, 272–274]. As there are  $\sim 10^3$  atoms per site, the critical value of  $(U/J)_c \approx 8 \times 10^3$  can only be achieved with an optical lattice of depth  $s = (1/4) \ln^2 \left(\frac{\sqrt{2}}{a_s q'} \left(\frac{U}{J}\right)_c\right) \approx 400$  (this expression is obtained by inversion of Eq. 4.61).

Experimentally the transition can be observed by looking at time-offlight images of the expanding gas. In the superfluid regime these should display several peaks that are related to quantized momentum states in the weakly interacting lattice. In this limit the momentum distribution has discrete momenta  $p_l = 2\pi\hbar l/d$  (l is an integer) [202], and corresponding expansion velocities  $v_l = l \times 0.126$  m/s, i.e. for expansion times of ~ 10 ms the peaks are already separated by ~ 1 mm. When increasing the lattice amplitude s the superfluid will become more localized, leading to decoherence between the individual lattice sites. In momentum space this can be observed by loss of the momentum peaks, resulting in a single broad momentum distribution in the center - the Mott insulator state. Using absorption imaging such a transition is not observed for a broad range of optical lattice powers, indicating that the Mott insulator state does not occur in the experiment.

#### Maintaining coherence

Although it is clear what happens in both  $U/J \ll 1$  and  $U/J \gg 1$ regimes, the lattice in our experiment is in the transition regime of  $U/J \approx 1$ . Some clear remarks about this regime are made in Figure 26.4 of Ref. [275]. In the region of  $U/J \approx 1$ , coherence among the lattice sites is still well maintained, whereas the number variance per lattice site becomes sub-Poissonian ('squeezed'). This can be understood as there has to be a transition from the coherent superfluid state with Poissonian number statistics to the highly correlated Mott insulator state, and in this transition region the onset of correlations leads to a sub-Poissonian distribution. As the spectroscopy beam addresses all lattice sites simultaneously in the experiment, the local number variance is not relevant as long as coherence (and therefore the mean field description) is maintained.

#### Does 2D scattering play a role?

One could make the argument that the BEC in the optical lattice would exist of a stack of independent 2D BEC 'pancakes' with modified *s*-wave scattering lengths [121]. This occurs in the limit where the 'pancakes' do not interact with each other. In our experimental regime - where the whole trap is superfluid - this does not play a role. The effective 2D scattering length scales with the ratio of the harmonic oscillator length of the lattice and the 3D s-wave scattering length as  $a_{2D} \propto e^{-a_{\text{latt}}/a_s}$  [121] which is very small  $(a_{2D} \approx 7 \times 10^{-22} \text{ m})$  as  $a_{\text{latt}}/a_s \approx 27$  in the experiment.

#### Interband spectroscopy

Based on the discussion of the various optical lattice effects in the previous sections, the BEC in the experiment can be described as a single BEC within the lowest band of the optical lattice situated at q' = 0. By absorbing a single photon from the spectroscopy laser the atoms gain a recoil  $q_{\rm spec} = 2\pi/\lambda_{\rm spec}$ , and they will acquire a quasimomentum  $q'_{\rm spec} = 2\pi \cos(\theta/2)/\lambda_{\rm spec}$  with respect to the optical lattice.

However, the experimental parameters are such that  $q'_{\text{spec}} = q$ . This means that excited atoms end up near the bandgap of the optical lattice as seen by the excited  $(2 \ ^1S)$  atoms. Due to the degeneracy of the lattice recoil and spectroscopy photon absorption recoil the experiment is always in this regime.

This agrees with the observation that the observed splitting between the two absorption lines can be manipulated by changing the relative polarization of the two beams as shown in Figure 4.3. The splitting does not disappear, which from an experimental point of view is caused by the fact that it becomes increasingly difficult to constrain the splitting if the two lineshapes overlap significantly. Second, there might not be a rotation angle for which the polarizations of both beams are orthogonal, meaning that it is impossible to completely remove the optical lattice effect.

It is useful to consider the scenario in which the largest optical lattice is applied that is shown Figure 4.3. The largest splitting achieved in the experiment is  $\Delta \approx 65$  kHz ( $\approx 3.25E_r$ ). Using  $\Delta/E_r = s/2$  (or looking at Figure 4.11), this splitting indicates that  $s \approx 6.5$ . This is the splitting as experienced by the 2 <sup>1</sup>S atoms (see Figure 4.2), which is proportional to the optical potential and therefore the polarizability of this state. As these atoms have a low exitation fraction during our spectroscopy and therefore a much lower density than the 2  ${}^{3}S$  state atoms, there is no other relevant macroscopic behavior caused by the 2  ${}^{1}S$  atoms apart from the observed bandgap in the lattice.

The ratio of the polarizabilities of the 2  ${}^{3}S$  and 2  ${}^{1}S$  states is -1.64 [133], so the optical lattice depth for the 2  ${}^{3}S$  atoms is  $s \approx 6.5/1.64 = 3.9$ . As this is an upper limit, it can be safely assumed that  $s \leq 3.9$  in all measurements. The experimental lower limit is achieved by trying to minimize the splitting with an estimated bandgap  $\Delta \approx 7.7$  kHz  $\approx 0.4E_r$  corresponding to  $s \approx 0.5$ . Although this is small, the 'swallow-tail' bandstructure regime that was briefly mentioned before is not reached as  $\mu = 0.21E_r < sE_r = 0.5E_r$ .

To summarize, from the measurements the lattice as observed by the BEC is estimated to be in the range 0.5 < s < 3.9 which is in the superfluid regime and the mean field description can still be used for the line shape model.

#### Deviations due to misalignment

The observed line splitting is caused by the fact that the lattice laser wavelength and spectroscopy laser wavelength are nearly degenerate and we derive some experimental parameters under the assumption that both wavelengths are equal and that the spectroscopy beam overlaps with the exact same angle as the optical dipole trap beam. The sensitivity of the calculations is estimated by assuming a worst-case scenario where the spectroscopy beam is fully aligned with the optical lattice (i.e. along the axial direction of the optical dipole trap), as this provides the largest difference between the lattice recoil energy and the spectroscopy photon recoil energy.

In this limit the recoil energies differ by  $E_r^{\rm s} - E_r \approx 0.6$  kHz =  $0.03E_r$  (see Table 4.2). This means that, in the band picture, the atom acquires a quasimomentum q' = 1.02q and therefore does not end up exactly at the bandgap. The band splitting near the bandgap has quasimomentum dependence (also see Eq. 4.58)

$$\frac{\Delta(q')}{E_r} = 2\sqrt{4\left(\frac{q'}{q} - 1\right)^2 + \frac{s^2}{16}},\tag{4.62}$$



**Figure 4.13** – Left: band splitting difference from bandgap  $\Delta(q' = q)$ near the bandgap for s = 3 (dashed), s = 4 (full), and s = 5 (dotted). Right: band splitting difference from bandgap  $\Delta(q' = q)$  for quasimomenta q' = 1.01q (dashed), q' = 1.02q (full), and q' = 1.03q (dotted).

which is displayed in Figure 4.13 as function of quasimomentum q' and the lattice amplitude s near q' = 1.02q. First of all the bandgap is minimal for q' = q. This means that if  $q' \neq q$ , the previous estimate will always be an upper limit (i.e. our lattice can only get smaller by correcting for this effect). Second, in the typical range of variation the corrections to the bandgap are  $< 0.01E_r$  i.e. below 1%. This is a worst-case estimate, and as the alignment and wavelengths are known to much better precision than this worst-case assumption, such deviations are negligible compared to the 2-digit accuracy in our analysis.

#### Concluding remarks about the optical lattice

In this appendix it is shown that the origin of the observed doublet in the BEC spectrum is understood by absorption of a spectroscopy photon recoil momentum in the presence of an optical lattice with a degenerate lattice (Bragg) recoil momentum. This is very similar to Bragg spectroscopy, but now between bands observed by two different electronic states [267].

The measured line splitting can be related to the lattice bandgap, and this gives an estimate of the optical lattice depth. For typical depths the lattice is relatively weak, meaning that the trapped BEC can be considered to be in the superfluid regime (as a single cloud instead of individual lattice sites) and not in the Mott insulator regime.

In the typical optical lattice regime, the chemical potential of the BEC is small enough to consider the BEC in the single-band regime i.e. the BEC is located around quasimomentum q' = 0 of the lowest band of the lattice. This means that the description of the BEC spectrum does not involve higher band contributions. Deviations from the degenerate case as presented here turn out to be negligible.

This phenomenon can present an opportunity to measure the polarizability of an excited state by observing the doublet splitting as function of the optical dipole trap wavelength. In the current experiment this is caused by an 'accidental' degeneracy, but an optical dipole trap wavelength and crossing angle can be chosen such that the resulting lattice recoil momentum is degenerate with the spectroscopy recoil momentum, thereby providing a method to measure polarizabilities over a broad range of wavelengths.

## 4.D Appendix: Extracting the *s*-wave scattering length

With the developed line shape model as discussed in Appendix 4.A, the Rabi frequency estimate from Appendix 4.B, and our understanding of the observed doublet in the Bose-Einstein condensate as discussed in Appendix 4.C, all the necessary information to use the line shape model is available to extract the  $2 {}^{3}S_{1} - 2 {}^{1}S_{0}$  s-wave scattering length  $a_{ts}$ . In this appendix the data analysis and results are discussed in more detail.

#### Acquiring the line shape data

The optical lattice amplitude is set such that the doublet splitting is  $\sim 35$  kHz, so the lines can be resolved individually and the Rabi frequency is large enough that each measurement can be done in a few seconds. The doublet is measured for six interaction times ranging from 0.5 s to 3.0 s, and the results are shown in Figure 4.14. Each line scan shows 100 data points which are normalized to 100 background measurements acquired in an alternating fashion during the measurement. The

results are represented by the chemical potential of the Bose-Einstein condensate normalized by their background level at the specific interaction time. The normalized data is represented using the chemical potential as this is the parameter for which Eq. 4.23 is solved. For ease of interpretation the data and line shape fits are rescaled such that they represent the atom number loss in Figure 4.4, but generally the data is analysed using the chemical potential representation.

#### Fitting procedure

For a dataset acquired with a spectroscopy laser interaction time t, the background chemical potential level is only determined for this time t. Using the average background chemical potential of all six datasets (i.e. for  $0.5 \text{ s} \leq t \leq 3.0 \text{ s}$ ) the one-body lifetime of the system can be constrained as a fixed parameter together with the chemical potential at t = 0 s, which is necessary for the line shape fitting. Sufficiently accurate estimates for the laser linewidth can be made by manual optimization (combined with the laser linewidth estimate from Section 2.5) in order to initialize the fitting optimization procedure.

For the fits as shown in Figure 4.14 the only free parameters are the absolute offset frequency, the doublet splitting, the relative amplitude between the two lines, and the factor  $[(a_{ts}/a_{ss}) - (\alpha_2/\alpha_1)]$  in which  $a_{ts}$  is the unknown s-wave scattering length. The fits of the six lines are performed simultaneously in *Mathematica* using a numerical optimization procedure which includes the statistical weight per datapoint *i* given as  $w_i = 1/\sigma^2$ , where  $\sigma$  is the propagated statistical uncertainty in the determined chemical potential (or atom number) per datapoint. The statistical uncertainty  $\sigma$  is based on analysis of the Gaussian distributed shot-to-shot fluctuations of the background level which are ~ 5%. This analysis does not take any uncertainties in the frequency axis into account, which are estimated to be  $\sigma_f \approx 1.8$  kHz predominantly by magnetic field fluctuations as determined by Van Rooij [190].

After the first optimization of the fit, the statistical weight per datapoint is reevaluated by including the frequency uncertainty into the signal uncertainty as  $\sigma_i^2 = \sigma^2 + (\partial \mu / \partial \nu)^2 \sigma_f^2$ , with  $\mu$  defined by Eqs. 4.2 and 4.23 and  $\partial \mu / \partial \nu$  is the sensitivity of the line shape model to the uncertainty in the frequency. With the updated statistical weights the numerical



**Figure 4.14** – Normalized chemical potential of the BEC doublet measured for interaction times ranging from 0.5 s (top left) to 3.0 s (bottom right) and simultaneous line shape fits (red lines). The bottom half of each plot shows the residuals including the error bars on the data, and the reduced  $\chi^2$  for the fit. 150

fitting procedure is reinitiated. The new resulting parameters have only slightly changed with respect to the first iteration (well within the initial evaluated uncertainties), but have acquired larger uncertainties as an additional error source is included. The results, including the residuals and the reevaluated error bars per datapoint, are shown in Figure 4.14. Based on this procedure  $\chi^2$  values are calculated and the scattering length  $a_{ts}$  is determined. The  $\chi^2$  values of the individual measurements can vary, but the  $a_{ts}$  values of all measurements are always in agreement with each other within their respective uncertainties.

#### Systematic effects in the line shape fits

As the line shape model depends on many parameters, the sensitivity of the scattering length  $a_{ts}$  as well as the  $\chi^2$  value of the fits is investigated for each individual parameter following a method where we vary a single parameter and consider the variation in the outcome. For parameters such as the initial chemical potential, lifetime, and laser linewidth, the sensitivity (i.e. relative change in  $a_{ts}$  for a relative change in the parameter) is approximately 19%, 1%, and 9% respectively. However, these parameters are known with sufficient accuracy to have a negligible contribution to the error budget compared to the ~ 25% uncertainty in the Rabi frequency, which is another parameter in the line shape model.

Within the fit parameter space limited by the experimental allowable bounds, only a single optimization of the total  $\chi^2$  of the six measurements could be found to provide a result on the scattering length  $a_{ts}$  as shown in Figure 4.15. Using standard  $\chi^2$  minimization routines [276] the total  $\chi^2$  as function of the scattering length  $a_{ts}$  can be fit with a parabola to determine the most likely value, and the second order derivative provides the statistical uncertainty in the determination of that value. The determined value of the scattering length is  $a_{ts} = +50(10)_{\text{stat}} a_0$ .

This result is obtained with a Rabi frequency which is within the estimated range of  $\Omega_R = 2\pi \times (21 \pm 5)$  Hz. Unfortunately the optimization could not be used to determine a more accurate value of the Rabi frequency as the scattering length is too sensitive to changes in the Rabi frequency (in other words:  $\chi^2$  varies only very little when varying the Rabi frequency). Therefore we consider the maximum variation of the Rabi frequency and see that the scattering length varies by  $\pm 43 a_0$  within



**Figure 4.15** – Optimization of the total  $\chi^2$  of all six line shape fits compared to the determined scattering length  $a_{ts}$ . The uncertainty in  $a_{ts}$  is given by the final uncertainty determined by the numerical fitting method as discussed in Section 4.D. Using a parabola to determine the scattering length for which  $\chi^2$  is minimized [276] we find  $a_{ts} = +50(10)_{stat} a_0$ , which is indicated by the dashed vertical line and the gray area representing the statistical uncertainty.

this range, whereas the total  $\chi^2$  changes by less than 1%. This range of scattering lengths is therefore the limiting systematic uncertainty.

We investigated the sensitivity of this result with all other parameters within their experimental range, and no changes beyond the quoted statistical error could be found. As a bonus a similar optimization of the expected laser linewidth around this minimum provided a linewidth of 4.5(3) kHz, which is in agreement with the estimates discussed in Chapter 2.

Based on the discussed optimization procedure the final result on the

scattering length is

$$a_{ts} = +50(10)_{\text{stat}}(43)_{\text{syst}} a_0. \tag{4.63}$$

#### Comparison to earlier mean field shift measurements

As a first comparison to the only other available data on the mean field shift, the value of the scattering length in Eq. 4.63 can be compared to the limits set by the mean field shift measurements done by Van Rooij [31, 190]. In that work the mean field shift was parametrized as

$$2\pi\Delta f_{\rm mf} = \frac{4\pi\hbar}{m} (a_{ts} - a_{ss})(n^{(2)} - n^{(1)}), \qquad (4.64)$$

where  $n^{(1)}$  and  $n^{(2)}$  are two different densities at which the transition frequency is measured. Such a measurement provides a limit on the scattering length differences as

$$|a_{ts} - a_{ss}| \le \frac{m}{2\hbar} \Big| \frac{\Delta f_{\rm mf}}{n^{(2)} - n^{(1)}} \Big|.$$
(4.65)

The mean field shift was determined to be zero within an uncertainty of 1.1 kHz. As the measurements were done at typical densities of  $3 \times 10^{13}$  cm<sup>-3</sup> and  $2 \times 10^{13}$  cm<sup>-3</sup>, this provides a limit on the scattering length difference range as

$$|a_{ts} - a_{ss}| \le 74 \ a_0, \tag{4.66}$$

or, using the known scattering length  $a_{ss} = 142.0(1) a_0 [152]$ ,

$$a_{ts} = +142(74) \ a_0. \tag{4.67}$$

As this range is based on the statistical uncertainty with which the mean field shift is determined, it should be interpreted as a  $1\sigma$  bound. As such, it is in agreement with our new determination although the accuracy has improved (even including the large systematic uncertainty). Furthermore, the new method depends on the actual line shape of the transition and not on an overall frequency shift.

#### Comparison to theory

Experimentally determined scattering lengths provide an excellent testing ground for molecular theory as the scattering lengths are sensitive to the exact molecular potential, and in particular to the energy of the least bound state in the potential [261]. For spin-stretched He<sup>\*</sup>-He<sup>\*</sup> collisions the measurement of binding energy of the least bound state in the  ${}^{5}\Sigma_{g}^{+}$  potential allowed a 0.07% accurate determination of the *s*-wave scattering length [152]. Equally impressive, the theoretical calculation provided a 0.3% accurate determination [144, 146] in good (2 $\sigma$ ) agreement. Due to its relative simplicity, He<sup>\*</sup>-He<sup>\*</sup> collisions are one of the few systems where the molecular potentials are known with such high accuracy [139, 140, 144, 146], and both theoretical and experimental work have been shown to be in good agreement when predicting for instance collision rates [141–143, 145, 153, 154] or interactions between He<sup>\*</sup> and alkali-atoms [159, 160].

There have not yet been any measurements on the  $2 {}^{3}S_{1} - 2 {}^{1}S_{0}$  molecular potential, as a mixture of these states is very unstable due to the 20 ms radiative lifetime of the  $2 {}^{1}S_{0}$  state and Penning ionization. However, this is not the first time that our group is interested in the molecular potential of these states as it once explained the large loss rate and narrow velocity distribution of metastable atoms for photo-assisted  $2 {}^{3}S_{1} - 2 {}^{3}P_{2}$  collisions [181]. At that time the presence of an avoided crossing between the  $1 {}^{3}\Sigma_{g}^{+}$  and  $2 {}^{3}\Sigma_{g}^{+}$  potentials (connecting to the  $2 {}^{3}S + 2 {}^{1}S$  and  $2 {}^{3}S + 2 {}^{3}P$  states, respectively) could explain the origin of fast escaping metastable atoms without requiring any calculation of the scattering length.

With the determined scattering length it is interesting to find out if a scattering length can be calculated from the *ab initio* molecular potentials [139]. We are fortunate to collaborate with Daniel Cocks and Ian Whittingham, who both have been involved with molecular potential calculations for He<sup>\*</sup> for quite some time [142, 143, 277–279]. Based on our communication with Daniel Cocks and Ian Whittingham [269], a brief overview of the method used to calculate the scattering lengths will be given.

As only s-wave collisions are allowed at in the ultracold regime ( $T \sim \mu K$ ), only molecular potentials with gerade symmetry are involved. The



**Figure 4.16** – Short-range behavior of the 1  ${}^{3}\Sigma_{g}^{+}$  (black) and 2  ${}^{3}\Sigma_{g}^{+}$  (red) potentials as used for the scattering length calculations with the long-range atomic states indicated. The data points taken from Ref. [139] and interpolated with a spline-based function. There is an avoided crossing at 9.5  $a_{0}$ , which is discussed in the text.

only relevant molecular potentials are then the  $1 \ {}^{3}\Sigma_{g}^{+}$  and  $2 \ {}^{3}\Sigma_{g}^{+}$  potentials, which are fitted using the long-range potential form

$$V_{\rm long}(R) = -\frac{C_6}{R^6} - \frac{C_8}{R^8} + E_{\rm asymp}$$
(4.68)

to determine the  $C_6$  and  $C_8$  coefficients. The asymptotic energy was fixed by the atomic energy levels [139]. The short-range potentials are based on tabulated data from Ref. [139] and shown in Figure 4.16. As can be seen in the figure, there is an avoided crossing between the potentials at an internuclear separation distance of ~ 9.5  $a_0$ . The coupling energy of the avoided crossing, defined as half the energy difference between the two potentials at 9.5  $a_0$ , is given in Ref. [139] as 18.5 meV, although variation of the coupling energy did not change the final results significantly. Penning ionization plays an important role in this molecular potential, and the total molecular potential has to include a loss channel. This is included by defining a complex interaction potential containing the calculated molecular potential and a so-called autoionization width (see e.g. [143]). As there is no information about the autoionization width for triplet-singlet collisions, the calculations assume a similar autoionization width as for the triplet-triplet collisions, as suggested in Ref. [139]. After calculating the wavefunctions in these complex potentials the complex scattering matrix can be calculated as function of the collision energy. Extrapolating the collision energy to zero this provides an *s*-wave

scattering length of  $a_{ts} = +42.45 \ a_0 - i \ 36.62 \ a_0$ . Based on 2% variations in the molecular potentials the recommended scattering length is [269]

$$a_{ts} = 42.5^{+0.5}_{-2.5} a_0 - i\,36.6^{+3.4}_{-2.1} a_0. \tag{4.69}$$

The scattering length has a large imaginary part caused by Penning ionization. As a result the real part of the scattering length is quite insensitive to variations in the molecular potentials which is the reason why it can be calculated with this high precision. In this regime, where the imaginary part is comparable to the real part, the complex interaction potential approach starts to break down but there is no theoretical framework yet which can circumvent this [269].

For elastic and inelastic collision rates both the real and imaginary part of the scattering length should be taken into account (see Appendix 5.A or Ref. [143]). But for the mean field interactions which are considered here the situation is more complicated as the real part of the scattering length is related to an energy shift and the imaginary part is related to a broadening effect [260]. Using the same model as given by Kokkelmans [260] and used in Chapter 3 (see Eq. 3.25), the broadening effect is estimated to be below 1 kHz. The mean field shift, which is defined in this model as a single frequency shift, is 3-4 kHz. This number cannot be simply compared with a shift in the measured line shapes as it is expressed in the asymmetry of the line shapes, but it is of the same order of magnitude and indicates that these numbers are realistic.

To summarize, from the complex interaction potentials using *ab initio* molecular potentials [139] Daniel Cocks and Ian Whittingham calculated a complex 2  ${}^{3}S - 2 {}^{1}S$  s-wave scattering length of  $a_{ts} = 42.5^{+0.5}_{-2.5} a_{0} -$ 

 $i 36.6^{+3.4}_{-2.1} a_0$ . The real part of this scattering length is related to the mean field shift as observed in our experiment, and is in agreement with the experimentally determined scattering length of  $a_{ts} = +50(10)_{\text{stat}}(43)_{\text{syst}} a_0$ .

# CHAPTER 5

### Towards a new measurement of the $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition

#### Abstract

For a sub-kHz accurate measurement of the  $2 {}^{3}S \rightarrow 2 {}^{1}S$  transition in helium not only the line shape of the transition, caused by quantum statistics and interactions, needs to be understood as was displayed in Chapter 4. Two systematic effects that limit the accuracy in frequency metrology work are the ac Stark shift and the Zeeman shift. In this Chapter we discuss how we can improve their measurement or even eliminate them.

#### 5.1 Introduction

With the improved linewidth of the spectroscopy laser the line shape of the transition has been resolved with sufficient precision to observe asymmetric features caused by the quantum statistics and interactions, as shown in Chapter 4. Although the line shapes contain interesting physics by themselves, understanding them and their associated systematic frequency shifts is only a single aspect in the next step for a

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sub-kHz accurate determination of the isotope shift on the 2  $^3S \rightarrow 2 \ ^1S$  transition frequency.

As with all high-precision experiments, the final result is, when the statistical error is small, only as good as its systematic error budget. In the final error budget by Van Rooij [190] the two dominating contributions to the error budget are the mean field shift (which is solved by resolving the asymmetric line shape), and the ac Stark shift extrapolation. The ac Stark shift is caused by the non-zero differential polarizability of  $2^{3}S$  and  $2^{1}S$  states at the optical dipole trap wavelength of 1557.3 nm. The solution for this non-zero differential polarizability is to find an alternative wavelength at which we can operate an optical dipole trap for which this effect is zero; a so-called 'magic wavelength'. This is not a new idea: in the optical clock community this is the only way to achieve the extreme accuracies that even go beyond the current definition of the SI second [227–229, 280, 281]. In this Chapter I will show how we calculated candidate magic wavelengths for the  $2^{3}S$  and  $2^{1}S$  states, and how we will implement this.

With the succesful implementation of a magic wavelength optical dipole trap the ac Stark shift no longer poses the dominant systematic uncertainty in the transition frequency determination, although there will be a remaining ac Stark shift from the spectroscopy laser. The next dominant systematic effect is the Zeeman shift on the transition. The Zeeman shift was previously measured with kHz accuracy, but with a standard deviation of  $\sim 2$  kHz on a daily basis [190] the uncertainty is too large for a sub-kHz absolute accuracy on the transition frequency. In this chapter I will discuss how we can improve on this measurement and perhaps even circumvent it.

#### 5.2 Magic wavelengths for the $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition in helium

In recent years a growing number of experimental tests of QED in atomic physics have surpassed the accuracy of theory, allowing new determinations of fundamental constants. High-precision spectroscopy in atomic hydrogen has been achieved with sufficient accuracy to allow a determination of the proton size from QED calculations [282], and spectroscopy in muonic hydrogen has allowed an even more accurate determination [94, 95]. Interestingly, the muonic hydrogen result currently differs by  $7\sigma$  from the proton size determined by hydrogen spectroscopy and electronproton collision experiments. So far there has not been a satisfying explanation for this discrepancy, which is aptly named the proton radius puzzle [97]. Research in this field has expanded to measurements in muonic helium ions, a hydrogenic system which has a different nuclear charge radius [100]. As this work is done for both naturally occurring isotopes of helium (<sup>4</sup>He and <sup>3</sup>He), the absolute charge radii of the  $\alpha$ particle and the helion may be determined at an aimed relative precision of  $3 \times 10^{-4}$  (0.5 attometer), providing a very interesting testing ground for both QED and few-body nuclear physics.

Parallel to these developments, high-precision spectroscopy in neutral helium has become an additional contribution to this field in recent years. Although QED calculations for three-body systems are not as accurate as for hydrogen(ic) systems, mass-independent uncertainties cancel when considering the isotope shift [103, 104]. Therefore isotopeshift measurements in neutral helium can provide a crucial comparison of the nuclear charge radius difference determined in the muonic helium ion and planned electronic helium ion measurements.

High-precision spectroscopy in helium is a well-established field, and transitions ranging from wavelengths of 51 nm to 2058 nm [29, 31, 33– 36, 44, 75–78] have been measured in recent years both from the ground state and from several (metastable) excited states. Only two transitions have been measured in both helium isotopes with sufficient precision for accurate nuclear charge radius difference determinations. The 2  ${}^{3}S \rightarrow$ 2  ${}^{3}P$  transition at 1083 nm [44] and the doubly-forbidden 2  ${}^{3}S \rightarrow$  2  ${}^{1}S$ transition at 1557 nm [31, 106] are measured at accuracies exceeding 10<sup>-11</sup>, providing an extracted nuclear charge radius difference with 0.3% and 1.1% precision, respectively. Interestingly, the determined nuclear charge radius differences from both experiments currently disagree by  $4\sigma$  [27, 44].

In order to determine the nuclear charge radius difference with a precision comparable to the muonic helium ion goal, we aim to measure the  $2 \ ^3S \rightarrow 2 \ ^1S$  transition with sub-kHz precision. One major improvement to be implemented is the elimination of the ac Stark shift induced by the optical dipole trap (ODT) in which the transition is measured. Many high-precision measurements involving optical (lattice) traps solve this problem by implementation of a so-called magic wavelength trap [229, 281]. In a magic wavelength trap the wavelength is chosen such that the ac polarizabilities of both the initial and final states of the measured transition are equal, thereby cancelling the differential ac Stark shift.

We calculate the wavelength-dependent (ac) polarizabilities of both metastable 2  ${}^{3}S$  and 2  ${}^{1}S$  states and identify wavelengths at which both are equal for either <sup>4</sup>He or <sup>3</sup>He. Generally one will find multiple magic wavelengths over a broad wavelength range, but our goal is to identify the most useful magic wavelength for our experiment. Currently [31, 35] we employ a 1557 nm ODT at a power of a few 100 mW, providing a trap depth of a few  $\mu$ K and a scattering lifetime<sup>1</sup> of > 100 s. For our future magic wavelength trap we need to produce a similar trap depth with sufficient laser power at that wavelength. Furthermore, the scattering rate should be low enough to have a lifetime of at least a few seconds, providing enough time to excite the atoms with a 1557-nm laser.

The purpose of this chapter is to show that it is possible to calculate magic wavelengths with sufficient accuracy to design an appropriate laser system solely based on *ab initio* level energies and Einstein A coefficients without having to resort to advanced theoretical techniques [134, 283]. Based on the calculations reported here, we are currently building a laser system at 319.82 nm with a tuning range of 300 GHz based on similar designs [284, 285].

The polarizabilities for the 2  ${}^{3}S$  and 2  ${}^{1}S$  states of <sup>4</sup>He are presented over a wavelength range from 318 nm to 2.5  $\mu$ m. In this range all magic wavelengths including estimated required ODT powers and corresponding trap lifetimes are calculated. From these results we identify our best candidate for a magic wavelength trap. A lot of work, both theoretical and experimental, has been done for the dc polarizability of the 2  ${}^{3}S$ and 2  ${}^{1}S$  states (see Table 5.1 for an overview). Therefore these are used as a benchmark for our calculations by also calculating the polarizabilities in the dc limit ( $\lambda \to \infty$ ). Calculations of the ac polarizability of the 2  ${}^{3}S$  and 2  ${}^{1}S$  states [135, 286] states allow for comparison of the

 $<sup>^1{\</sup>rm The}$  actual lifetime in the trap is limited to 10's of seconds due to background collisions.

polarizability calculations at finite wavelengths.

Finally we present a simple extension to <sup>3</sup>He which has a hyperfine structure that needs to be taken into account. Although different theoretical challenges arise due to the hyperfine interaction, we can get an estimation of the <sup>3</sup>He magic wavelength candidates and show that they are equal to the <sup>4</sup>He results approximately shifted by the hyperfine and isotope shift.

#### Theory for <sup>4</sup>He

For an atomic state with angular momentum J and magnetic projection  $m_J$ , the polarizability  $\alpha$  induced by an electromagnetic wave with polarization state q ( $q = 0, \pm 1$ ) and angular frequency  $\omega$  due to a single opposite parity state is [287]

$$\alpha^{(n)}(J, m_J, J', m'_J, q) = 6\pi\epsilon_0 c^3 (2J'+1) \\ \times \begin{pmatrix} J & 1 & J' \\ -m_J & q & m'_J \end{pmatrix}^2 \frac{A_{nJJ'}}{\omega_{nJJ'}^2 (\omega_{nJJ'}^2 - \omega^2)}.$$
 (5.1)

Here  $\omega_{nJJ'}$  is the 2 <sup>1,3</sup> $S_J \rightarrow n$  <sup>1,3</sup> $P_{J'}$  transition frequency and  $A_{nJJ'}$  the Einstein A coefficient of the transition. The term between two brackets represents the 3*j* symbol of the transition. The total polarizability  $\alpha(J, m_J, q)$  is given by a sum over all opposite-parity states as

$$\alpha(J, m_J, q) = \sum_n \sum_{J'} \alpha^{(n)}(J, m_J, J', m'_J, q).$$
 (5.2)

In a general way the polarizability  $\alpha$  can be written as the sum of a scalar polarizability, independent of  $m_J$ , and a tensorial part describing the splitting of the  $m_J$  levels [283, 288]. Within the LS coupling scheme the tensor polarizability of the 2  ${}^3S_1$  and 2  ${}^1S_0$  states in  ${}^4\text{He}$  is zero and the polarizability is defined by averaging over all  $m_J$  states and therefore independent of  $m_J$ . As our experimental work specifically concerns the spin-stretched 2  ${}^3S_1$  ( $m_J = +1$ ) state [31, 35], Eqs. 5.1 and 5.2 are used to calculate the polarizability for the  $m_J = +1$  state assuming linearly polarized light (q = 0). For  ${}^3\text{He}$  the calculations specifically concern the spin-stretched 2  ${}^3S_1$  ( $F = 3/2, m_F = +3/2$ ) and 2  ${}^1S_0$  ( $F = 1/2, m_F = +1/2$ ) states.

The higher-order contribution to the Stark shift, the hyperpolarizability, is estimated using calculations of a similar system [289]. The contribution is many orders of magnitude smaller than the accuracy of our calculations and therefore neglected.

The summation in Eq. 5.2 can be explicitly calculated for  $2^{1,3}S \rightarrow n^{1,3}P$  transitions up to n = 10, as accurate *ab initio* energy level data and Einstein A coefficients are available [56]. Extrapolation of both the energy levels and the Einstein A coefficients is required to calculate contributions of dipole transition matrix elements with states beyond n = 10. A straightforward quantum defect extrapolation can be used to determine the energies using the effective quantum number  $n^*$  [290]:

$$n^* = n - \sum_{r=0}^{\infty} \frac{\delta_r}{n^{*r}},\tag{5.3}$$

where  $\delta_r$  are fit parameters and the quantity  $n-n^*$  is commonly referred to as the quantum defect. For both the singlet and triplet series, Eq. 5.3 is used to fit the literature data up to n = 10 and to extrapolate to arbitrary n. This method is tested using a dataset provided by Drake [290].

Extrapolation of the Einstein A coefficients is more complicated as there is no relation such as Eq. 5.3 for Einstein A coefficients. Furthermore, the sum-over-states method does not provide straightforward extrapolation beyond the ionization limit, as the energy levels converge to the ionization limit for  $n \to \infty$ . Both problems can be solved by calculating the polarizability contribution of a single transition  $2 {}^{3}S_{1} \to n {}^{3}P_{J'}$ (or  $2 {}^{1}S_{0} \to n {}^{1}P_{1}$ ) as given in Eq. 5.2 and defining the polarizability density per upper state energy interval as

$$\frac{\Delta \alpha^{(n)}}{\Delta E} = \frac{2\alpha^{(n)}}{E_{n+1} - E_{n-1}},$$
(5.4)

which is evaluated at  $E_n$ .  $E_{n+1}$  and  $E_{n-1}$  are the energies of the neighbouring upper states with the same value of J'. The energies are given by the Rydberg formula  $E_n(n^*) = E_{\rm IP} - R_{\infty}/n^{*2}$ , where  $E_{\rm IP}$  is the ionization potential of the ground state. For ease of notation we have omitted all the dependent variables of  $\alpha^{(n)}$  as defined in Eq. 5.1. The polarizability density is a function of energy and can not only be used to calculate the polarizability contribution from dipole transition matrix elements to highly excited (Rydberg) states, but additionally allows extrapolation beyond the ionization potential. Using the Rydberg formula, the polarizability density becomes

$$\frac{\Delta \alpha^{(n)}}{\Delta E} = \frac{\alpha^{(n)}}{R_{\infty}} \frac{(n^{*2} - 1)^2}{2n^*},$$
(5.5)

where we have made the approximation that  $n - n^*$  is constant for increasing n. This approximation already works better than 1% for n = 2. In the limit  $n \gg 1$ , the polarizability contribution per energy interval can be written as

$$\frac{d\alpha^{(n)}}{dE} = \frac{6\pi\epsilon_0 c^3}{R_\infty} (2J'+1) \begin{pmatrix} J & 1 & J' \\ -M_J & q & M'_J \end{pmatrix}^2 \frac{C_{nJJ'}(n^*)}{\omega^2_{nJJ'}(\omega^2_{nJJ'}-\omega^2)}, \quad (5.6)$$

where we define

$$C_{nJJ'}(n^*) \equiv \frac{A_{nJJ'}(n^{*2}-1)^2}{2n^*}.$$
(5.7)

As there is no exact analytical model for  $A_{nJJ'}$  as function of energy, the method of extrapolation is based on a simple low-order polynomial fit of the  $C_{nJJ'}(n^*)$  as function of  $E(n^*)$  for the  $n \leq 10$  levels. The result is a function  $C_{nJJ'}(E)$  that is used to extrapolate  $A_{nJJ'}$  to arbitrary upper states and calculate the corresponding polarizability contributions. This method can be used to calculate the finite polarizability contributions of all Rydberg states for  $n \to \infty$ . As the general behavior of the Einstein A coefficients is proportional to  $n^{*-3}$  for the Rydberg states,  $C_{nJJ'}(E)$ will have a finite value at the ionization potential indicating that contributions from the continuum have to be taken into account as well. As the extrapolation is a function of energy, it is extended beyond the ionization potential to calculate additional continuum contributions to the polarizability. This omits all higher order effects such as resonances to doubly-excited states or two-photon excitations into the continuum, and it should be considered as an approximation of the continuum.

For a large enough quantum number n, the discrete sum-over-states method smoothly continues as an integration-over-states method following Eq. 5.6. The ionization potential serves as a natural choice as the energy at which the calculation would switch from the discrete sum to the integration method. But even for large enough n there is a negligible numerical error in varying the exact cutoff energy  $E_c$  at which we switch between these methods. The calculation of the total polarizability is therefore performed using the sum-over-states method to an arbitrary cutoff at  $E_c = E_{\rm IP} - R_{\infty}/n_{max}^{*2}$  and continued with an integration over the remaining states as

$$\alpha^{\text{cont}}(J, m_J) = \sum_{J'} \int_{E_c}^{\infty} \frac{d\alpha^{(n)}}{dE} dE, \qquad (5.8)$$

where E is the energy of the corresponding state. A low-order polynomial fit of Eq. 5.7 is used to calculate  $d\alpha^{(n)}/dE$  such that the integral of Eq. 5.8 provides an analytical solution. The total polarizability is therefore easily calculated as a sum-over-states part and an analytical expression

$$\alpha(J, m_J) = \alpha^{\text{cont}} + \sum_{n=1}^{n=n_{max}} \sum_{J'} \alpha^{(n)}.$$
(5.9)

#### Numerical uncertainties

In this section we discuss the sources of any numerical errors in our calculations, which are purely based on the technical execution of our method. The accuracy of our calculations due to our estimation of the continuum contribution will be discussed in Section 5.2 where our results are compared to other calculations.

The numerical convergence of Eq. 5.9 is tested by varying  $n_{max}$ . The polarizability converges as  $n_{max}^{-2}$  and even for  $n_{max} = 20$  the polarizability is within a fraction  $10^{-4}$  of the polarizability calculated using  $n_{max} = 5000$ . The computation of Eq. 5.9 therefore poses no numerical problems.

A more crucial matter is the fact that our calculations are based on two extrapolations: that of the level energies and the Einstein A coefficients. For the  $n \leq 10$  levels in helium the *ab initio* calculations of the level energies and Einstein A coefficients are used [56]. The higher level energies are extrapolated using Eq. 5.3 and include up to fifth order (r = 5) contributions. Variation of the total number of orders (r = 4, 6) or using a different dataset (such as the NIST database [291] as used in other recent work [135]) affects the polarizabilities at the  $10^{-8}$  level and is negligible.

The limiting factor in the accuracy of the calculations is the choice of extrapolation of the Einstein A coefficients through extrapolation of  $C_{nJJ'}(E)$ . As mentioned before, no advanced methods are used to calculate transition matrix elements to higher states or doubly excited states in the continuum. The heuristic approach we use instead, is to choose an extrapolation function that is smooth, continuous and provides a convergent integral in Eq. 5.8. A number of different functions have been tried which provide a similar quality of the fit, and their effect on the calculation of the continuum contribution can lead to a polarizability shift which is a significant fraction of the continuum contribution itself. In our calculations this is the limiting factor in the accuracy of the calculated magic wavelengths. A second order polynomial function is chosen to extrapolate  $C_{nJJ'}(E)$  as it has the additional advantage of providing an analytical solution of the continuum contributions.

The absolute accuracy of the calculations will be discussed the next section and determines the accuracy given in the calculated magic wavelengths later in this chapter.

#### **Results - dc polarizabilities**

In order to discuss the absolute accuracy of the calculations, we first present our polarizabilities calculated in the dc limit  $(\lambda \to \infty)$  as a lot of literature is available for these calculations. After comparison with the dc polarizabilities, the ac polarizabilities are given in the next section including the magic wavelengths at which they are equal for the 2  ${}^{3}S_{1}$   $(m_{J} = +1)$  and 2  ${}^{1}S_{0}$  states. Experimental characteristics, such as the required trapping power and scattering lifetime at the magic wavelengths, are estimated in order to discuss which magic wavelength candidate is most suitable for our experiment. After that the tune-out wavelength (where the polarizability is zero) of the 2  ${}^{3}S_{1}$  state near 414 nm is compared to the result calculated by Mitroy and Tang [135]. An overview of previously calculated and measured dc polarizabilities for the 2  ${}^{1}S_{0}$  and 2  ${}^{3}S_{1}$  states of <sup>4</sup>He is given in Table 5.1 together with

**Table 5.1** – Comparison of calculations and measurements of  $m_J$ -averaged dc polarizabilities of the 2  ${}^1S_0$  and 2  ${}^3S_1$  states in units of  $a_0^3$ .

Author (year)	Ref.	$2 \ {}^1S_0$	$2 \ {}^{3}S_{1}$
Crosby and Zorn (1977) [Exp.]	[292]	729(88)	301(20)
Ekstrom $et al.$ (1995) [Exp.]	[293, 294]		322(6.8)
Chung and Hurst $(1966)$	[295]	801.95	315.63
Drake (1972)	[296]	800.2	315.608
Chung (1977)	[297]	801.10	315.63
Glover and Weinhold (1977)	[298]	803.31	316.24
Lamm and Szabo (1980)	[299]	790.8	318.7
Bishop and Pipin (1993)	[300]		315.631
Rérat <i>et al.</i> (1993)	[301]	803.25	
Chen (1995)	[286]	800.31	
Chen and Chung (1996), $B$ spline	[302]		315.630
Chen and Chung (1996), Slater	[302]		315.611
Yan and Babb (1998)	[134]	800.31666	315.631468
Mitroy and Tang (2013), hybrid	[135]		315.462
Mitroy and Tang (2013), CPM	[135]		316.020
This work		801.19	317.64

our results. For convenience the polarizabilities are given in atomic units  $a_0^3$  ( $a_0$  is the Bohr radius), but they can be converted to SI units through multiplication by  $4\pi\epsilon_0 a_0^3 \approx 1.64877 \times 10^{-41} \text{ JV}^{-2}\text{m}^2$ . Furthermore, the dc polarizabilities are calculated using the common convention of averaging over all  $m_J$  states and all possible polarizations q [283].

There is general agreement between our results and previously calculated dc polarizabilities, but comparison with the work of Yan and Babb [134], which provides the most accurate calculated dc polarizabilities to date, shows that both our 2  ${}^{1}S_{0}$  and 2  ${}^{3}S_{1}$  dc polarizabilities are slightly larger (0.1% and 0.6%, respectively). The difference is comparable to the uncertainty in the calculated continuum contributions as discussed before, and we conclude that our absolute accuracy is indeed limited by the exact calculation of the continuum contributions. It should be **Table 5.2** – Calculated magic wavelengths  $\lambda_m$  for the 2  ${}^3S_1$  $(m_J = +1) \rightarrow 2 {}^1S_0$  transition with the corresponding differential polarizability slope  $d\alpha/d\lambda$  and the absolute polarizability  $\alpha$  at the magic wavelength. The last row gives the wavelength and polarizability at which we currently use our ODT. Additional columns give the laser beam power required to create a 5  $\mu K$  deep trap in the exact same crossed-beam geometry as currently employed and the corresponding lifetime of the gas in this geometry due to scattering from a nearby 2  ${}^3S_1 \rightarrow n {}^3P_{0,1,2}$  transition. See Appendix 5.2 for details on those calculations.

$\lambda_m \text{ [nm]}$	$dlpha/d\lambda$	$\alpha [a_0^3]$	Laser	Lifetime	Nearest
	$[a_0^3/{\rm nm}]$	- •-	power $[W]$	$[\mathbf{s}]$	transition
318.611	$-7.00 \times 10^4$	-809.2			
319.815	$-4.40  imes 10^3$	189.3	0.7	3	$2 \ {}^{3}S_{1}$ -4 $ {}^{3}P$
321.409	$-5.38  imes 10^2$	55.3	2.3	6	$2 {}^{3}S_{1}$ - $4 {}^{3}P$
323.587	$-1.48 \times 10^2$	17.2	7.3	6	$2 \ {}^{3}S_{1}$ -4 $ {}^{3}P$
326.672	$-5.48 \times 10^1$	-1.2			
331.268	$-2.37  imes 10^1$	-13.5			
338.644	$-1.08  imes 10^1$	-24.2			
352.242	-5.33	-39.0			
411.863	-2.00	4.5	28.0	4	$2 \ {}^{3}S_{1}$ -3 ${}^{3}P$
1557.3	0.0	603.8	0.2	205	$2 {}^{3}S_{1}$ - $2 {}^{3}P$

noted that the continuum contributions in the dc limit are 7.1  $a_0^3$  and 3.6  $a_0^3$ , respectively. This only contributes 1% to the total polarizability in contrast to e.g. ground-state hydrogen for which the continuum contribution is 20% of the total polarizability [303].

#### **Results - magic wavelengths**

We have calculated the ac polarizabilities of the 2  ${}^{1}S_{0}$  and 2  ${}^{3}S_{1}$  $(m_{J} = +1)$  states in the range of 318 nm to 2.5  $\mu$ m and an overview of the identified magic wavelengths is shown in Table 5.2. The slope of the differential polarizability is also given in order to estimate the sensitivity of the determined magic wavelength due to the accuracy of the calcu-



**Figure 5.1** – Calculated polarizabilities of the 2  ${}^{3}S_{1}$  (dashed, blue) and 2  ${}^{1}S_{0}$  (dotted, black) states shown together with the differential polarizability (full, red) in the wavelength range 318-327 nm. The blue and black vertical lines indicate the positions of the 2  ${}^{3}S_{1} \rightarrow 4 {}^{3}P$  and the 2  ${}^{1}S_{0} \rightarrow n {}^{1}P$  (n = 9 - 13) transitions, respectively. There are five magic wavelengths (black dots) in this range, all listed in Table 5.2.

lated polarizabilities. Table 5.2 furthermore provides the trapping beam power required to produce a trap depth of 5  $\mu$ K and the corresponding scattering lifetime to indicate the experimental feasibility of each magic wavelength.

The magic wavelengths in the range 318-327 nm, as shown in Figure 5.1, are mainly due to the many resonances in the singlet series. The most promising magic wavelength for application in the experiment is at 319.815 nm, as the polarizability is large enough to provide sufficient trap depth at reasonable laser powers while the estimated scattering lifetime is still acceptable (see Table 5.2).

The magic wavelengths at 318.611 nm and 326.672 nm are not useful for our experiment as the absolute  $2 {}^{3}S_{1}$  polarizability is negative and therefore a focused laser beam does not provide a trapping potential. There are more magic wavelengths for  $\lambda < 318.611$  nm, but the polarizability



**Figure 5.2** – Calculated polarizabilities of the 2  ${}^{3}S_{1}$  (dashed, blue) and 2  ${}^{1}S_{0}$  (dotted, black) states shown together with the differential polarizability (full, red) for wavelengths ranging from 327 nm to 420 nm. The blue and black vertical lines indicate the positions of the 2  ${}^{3}S_{1} \rightarrow 3 {}^{3}P$  and the 2  ${}^{1}S_{0} \rightarrow n {}^{1}P$  (n = 4 - 8) transitions, respectively. There are four magic wavelengths (black dots) in this range, all listed in Table 5.2. The inset shows the wavelength region 411-415 nm, displaying the magic wavelength at 411.863 nm and the tune-out wavelength of the 2  ${}^{3}S_{1}$  state at 414.197 nm.

of the 2  ${}^{3}S_{1}$  state will stay negative until the ionization wavelength of the 2  ${}^{1}S$  state around 312 nm. In the range 327-420 nm, shown in Figure 5.2, there are four more magic wavelengths. The magic wavelength at 411.863 nm, previously predicted with nm accuracy [76], is the only one in this region with a small yet positive 2  ${}^{3}S_{1}$  polarizability (see inset in Figure 5.2). There are no more magic wavelengths in the range 420 nm-2.5  $\mu$ m, which is shown in Figure 5.3, and the polarizabilities converge to the dc polarizabilities for  $\lambda > 2.5 \mu$ m. The ac polarizability of the 2  ${}^{1}S_{0}$  state can be compared to previous polarizability calculations from dc to 506 nm [286]. Combined with the dc polarizability comparison and the tune-out wavelength result for the 2  ${}^{3}S_{1}$  state, we find that the



**Figure 5.3** – Calculated polarizabilities of the 2  ${}^{3}S_{1}$  (dashed, blue) and 2  ${}^{1}S_{0}$  (dotted, black) states shown together with the differential polarizability (full, red) for wavelengths ranging from 420 nm to 2.5  $\mu$ m. The blue and black vertical lines indicate the positions of the 2  ${}^{3}S_{1} \rightarrow 2 {}^{3}P$  and the 2  ${}^{1}S_{0} \rightarrow n {}^{1}P$  (n = 2, 3) transitions, respectively. There are no magic wavelengths in this range and the polarizabilities converge to the dc polarizabilities for  $\lambda > 2.5 \ \mu$ m.

accuracy of our calculations is limited by the exact calculation of the continuum contributions. We note that around 320 nm the absolute continuum contributions (26  $a_0^3$  and 5.5  $a_0^3$  for the 2 <sup>1</sup>S and 2 <sup>3</sup>S states, respectively) and the corresponding uncertainty have increased, as the shorter wavelengths are closer to the 2 <sup>1</sup>S ionization limit at 312 nm. The uncertainty in the absolute value of the polarizabilities translates to an uncertainty in the absolute value of the magic wavelength through the slope  $d\alpha/d\lambda$  of the differential polarizability at the zero crossing. For the magic wavelength at 319.815 nm this gives a frequency uncertainty of 10 GHz (0.003 nm), yet for the magic wavelength near 412 nm the uncertainty is approximately 1 nm due to the very small slope at the zero crossing. However, the latter magic wavelength is not suitable for our experiment as the absolute polarizability is very small.

#### Tune-out wavelength of the 2 $^{3}S_{1}$ state

The zero crossings of the absolute polarizability of a single state occur at so-called tune-out wavelengths. Mitroy and Tang calculated several tune-out wavelengths for the 2  ${}^{3}S_{1}$  state [135], of which the candidate at 413.02 nm is the most sensitive to the absolute value of the polarizability due to a very small slope at the zero crossing. We find this tune-out wavelength at 414.197 nm (see inset in Figure 5.2), which is considerably larger. However, the slope of the polarizability at the zero crossing can be used to calculate that the difference in tune-out wavelength is equivalent to a difference in the calculated absolute polarizabilities. Comparison of the calculated dc polarizabilities (see Table 5.1) shows a similar difference, so within a constant offset of the absolute polarizability our tune-out wavelength is in agreement with Mitroy and Tang's result.

#### Extension to <sup>3</sup>He

The  $2 {}^{3}S \rightarrow 2 {}^{1}S$  transition is also measured in <sup>3</sup>He in order to determine the isotope shift of the transition frequency [31]. Hence a magic wavelength trap for <sup>3</sup>He will be required as well. As <sup>3</sup>He has a nuclear spin (I = 1/2), the measured hyperfine transition is  $2 {}^{3}S$   $(F = 3/2, m_F =$  $+3/2) \rightarrow 2 {}^{1}S$   $(F = 1/2, m_F = +1/2)$  and the magic wavelengths need to be calculated for these two spin-stretched states.

The mass-dependent (isotope) shift of the energy levels is taken into account by using <sup>3</sup>He energy level data [30] and recalculating the quantum defects using Eq. 5.3. The Einstein A coefficients of the transitions also change due to the different reduced mass of the system [56], but this effect is negligible compared to the accuracy of the calculations. In total, the mass-dependent shift of the magic wavelengths is dominated by the shift of the nearest transitions and is approximately -45 GHz.

The fine-structure splitting decreases as  $1/n^3$  whereas the hyperfine splitting converges to a constant value for increasing n [304]. In this regime the (LS)JIF coupling scheme is not the best coupling scheme because J is no longer a good quantum number. Instead an alternative coupling scheme is used which first couples the nuclear spin quantum number I and total electron spin S to a new quantum number K [305]. This new quantum number K then couples to L to form the total angular momentum F. In this coupling scheme the transition strengths can be calculated with better precision compared to the (LS)JIF coupling scheme, and can be applied for states with  $n \ge 3$ . Although this coupling scheme does not work perfectly for n = 2 (which in any case is far-detuned from the magic wavelengths), it provides an estimate of the transition strengths that is sufficiently accurate for our purposes.

For increasing n, the strong nuclear spin interaction with the 1s electron becomes comparable with the exchange interaction between the 1s and np electrons [304]. This leads to mixing of the singlet and triplet states as the total electron spin S is no longer a good quantum number. The solution requires exact diagonalization of the Rydberg states, which provides the singlet-triplet mixing and the energy shifts of the states. The mixing parameter is then used to correct the Einstein A coefficients and the energies of the states. Although this is implemented in the calculations, these corrections lead to shifts in the magic wavelengths that are below the absolute accuracy of the calculations.

Due to the two hyperfine states of  ${}^{3}\text{He}^{+}$  in the 1s ground state, there are two Rydberg series in the  ${}^{3}\text{He}$  atom. For even higher n than discussed before, this leads to mixing of Rydberg states with different n [304]. The resulting shifts in the polarizabilities are well below the accuracy of the calculations and are therefore neglected.

Using the aforementioned adaptations, the polarizability of the 2  ${}^{3}S$   $(F = 3/2, m_F = +3/2)$  and 2  ${}^{1}S$   $(F = 1/2, m_F = +1/2)$  states can be calculated using Eq. 5.1, but with substituted quantum numbers  $(J, m_J \rightarrow F, m_F)$ , Einstein A coefficients and transition frequencies. The numerical calculation of the polarizabilities and discussion of the numerical accuracies is similar to the <sup>4</sup>He case. An additional uncertainty of 1.0  $a_0^3$  is added in the calculation of the polarizabilities of the <sup>3</sup>He states based on a conservative estimate of the shifts caused by the hyperfine interaction. It should be noted that the states of interest, 2  ${}^{1}S$  and 2  ${}^{3}S$ , both have angular momentum L = 0 and both are in the fully spin-stretched state. Therefore neither  ${}^{3}$ He nor  ${}^{4}$ He has a tensor polarizability for the states discussed in this paper.

A comparison between the  ${}^{4}$ He and  ${}^{3}$ He magic wavelengths is presented in Table 5.3. Magic wavelengths up to 330 nm are all shifted by the isotope shift with small corrections due the abovementioned effects. The frequency difference between the two isotopes (third column of Table
**Table 5.3** – Comparison of magic wavelengths  $\lambda_m$  calculated for the <sup>4</sup>He 2  ${}^{3}S_1 (m_J = +1) \rightarrow 2 {}^{1}S_0$  and  ${}^{3}He 2 {}^{3}S_1 (F = 3/2, m_F = +3/2) \rightarrow 2 {}^{1}S_0 (F = 1/2, m_F = +1/2)$  transitions and the corresponding frequency shift. The uncertainty in the shift is due to the additional 1.0  $a_0^3$  absolute uncertainty in the polarizabilities of  ${}^{3}He$ .

$\lambda_m \text{ [nm]}$		Shift [GHz]	
$^{4}\mathrm{He}$	$^{3}\mathrm{He}$		
318.611	318.626	-45.03(4)	
319.815	319.830	-43.1(7)	
321.409	321.423	-38(5)	
323.587	323.602	$-4(2) \times 10^1$	

5.3) grows with increasing wavelengths because  $d\alpha/d\lambda$  decreases and the results become more sensitive to the absolute accuracy  $(1.0 \ a_0^3)$  of the calculations, as can be seen from the growing uncertainties associated with the shifts. The isotope shifts for magic wavelengths with  $\lambda > 324$  nm have been omitted in Table 5.3 as they are not useful due to the large relative uncertainty.

The difference of the magic wavelengths between the two isotopes is well within the tuning range of our designed laser system near 320 nm. Furthermore there is no significant change in the absolute polarizability or the slope  $d\alpha/d\lambda$  at the magic wavelengths. This means that an ODT at these wavelengths has a comparable performance for either isotope.

#### Summary and outlook

We have calculated the dc and ac polarizabilities of the 2  ${}^{1}S$  and 2  ${}^{3}S$  states for both <sup>4</sup>He and <sup>3</sup>He in the wavelength range of 318 nm to 2.5  $\mu$ m and determined the magic wavelengths at which these polarizabilities are equal for either isotope. The accuracy of our simple method is limited by the extrapolation of the polarizability contributions of the continuum states. This is less than achievable through more sophisticated methods which calculate the transition matrix elements explicitly. However, the purpose of this work is to show that using a simple extrapolation method

it is possible to achieve an accuracy on the order of 10 GHz for the magic wavelengths that are of experimental interest, which is required to design an appropriate laser system for the required wavelengths.

Most experimentally feasible magic wavelength candidates are in the range of 319-324 nm, as the absolute polarizability of the 2  ${}^{3}S_{1}$  state in this range is positive and large enough to create reasonable ( $\sim \mu K$ ) trap depths in a crossed-beam ODT with a few Watts of laser power. The estimated scattering rates at these wavelengths and intensities are low enough to perform spectroscopy on the doubly-forbidden 2  ${}^{3}S \rightarrow 2 {}^{1}S$  transition.

The calculations are extended to also calculate magic wavelengths in <sup>3</sup>He. Although the hyperfine structure, which is absent in <sup>4</sup>He, leads to complications in the calculation of the polarizabilities, these effects are very limited for the 2 <sup>1</sup>S and 2 <sup>3</sup>S states. The magic wavelengths of interest, around 320 nm, are shifted relative to the <sup>4</sup>He magic wavelengths by predominantly the isotope shift.

#### **Recent developments**

Briefly after publication [133], the tune-out wavelength around 414 nm was measured with  $5 \times 10^{-6}$  relative accuracy [136] and found to be in agreement with our result. The experimental result is also in full agreement with the extensive calculation by Mitroy and Tang [135], confirming our observation of a small systematic offset in the absolute polarizability which is within the total uncertainty of our calculation. Since November 2014 the UV laser setup was constructed and produced up to 2 W of UV light, which was more than sufficient to optically trap <sup>4</sup>He<sup>\*</sup> atoms [198]. From these measurements the heating and scattering rates of the gas are determined to be low enough to trap atoms long enough to be interrogated for high-precision spectroscopy. Currently atoms have been loaded in a full crossed-beam UV optical dipole trap and initial measurements for spectroscopy are ongoing.

#### Appendix: Trap depth and lifetime calculations

An overview of optical dipole traps and the equations used in this Appendix can be found in Section 2.1. The depth U of a crossed-beam ODT, as currently used in our experiment [31, 35], is

$$U = 2\frac{\alpha}{2\epsilon_0 c} \frac{2P}{\pi w_0^2},\tag{5.10}$$

where  $\alpha$  is the polarizability of the 2  ${}^{3}S_{1}(m_{J} = +1)$  state, P the power of the incident trapping laser beam and  $w_{0}$  the beam waist. In our experiment, the first ODT beam is reused by refocusing it through the original focus ( $w_{0} \approx 85 \ \mu m$ ) at an angle of 19° with respect to the original beam. At the currently used ODT wavelength of 1557.3 nm the polarizability is  $\alpha = 603.8 \ a_{0}^{3}$  (see Table 5.2) which gives a trap depth of approximately 5  $\mu K$  at an ODT beam power of  $P = 210 \ mW$ . In Table 5.2 we use Eq. 5.10 to calculate the trapping power at the different magic wavelengths corresponding to a trap depth of 5  $\mu K$  to indicate the required beam power that should be produced at that magic wavelength.

As a good approximation of the lifetime of the atoms in the ODT due to scattering, one can take the nearest transition into account to calculate the corresponding scattering rate. The scattering rate  $\Gamma_{sc}$  is

$$\Gamma_{\rm sc} = \frac{6\pi c^2 \omega^3}{\hbar} \left( \frac{\Gamma}{\omega_0^2 (\omega_0^2 - \omega^2)} \right)^2 I_0, \tag{5.11}$$

where  $I_0$  is the total intensity of the light,  $\omega$  the angular frequency of the trapping light and  $\omega_0$  and  $\Gamma$  the transition frequency and linewidth (all in rad s<sup>-1</sup>). The nearest transitions are given in Table 5.2, and the lifetime  $1/\Gamma_{\rm sc}$  is given for each magic wavelength trap using the required trapping beam power calculated to provide a 5  $\mu$ K deep trap.

# 5.3 Ramsey-type Zeeman spectroscopy in He<sup>\*</sup>

In the previous measurement of the doubly forbidden 2  ${}^{3}S \rightarrow 2 {}^{1}S$ transition [31, 190], contributions to the systematic error budget came from the ac Stark shift extrapolation, mean field shift, and the Zeeman shift. As discussed in the previous section, the ac Stark shift will be reduced by implementing a magic wavelength trap [133, 198] and the mean field shift can be extracted from the exact line shape measurements (Chapter 4). The Zeeman shift, which was limiting the error budget total of 1.8 kHz by 0.5 kHz, needs to be measured with higher accuracy than before.

The Zeeman shift was previously measured by rf spectroscopy of the Zeeman splitting of the  $m_J$  states of <sup>4</sup>He<sup>\*</sup>. Typically such measurements reached kHz-accuracy, but the method depends on fitting a resonance peak of which the resonance position is very sensitive to the background noise. Additionally it turns out that mean field interactions between the different magnetic substates play a non-negligible role. We thus require a different method which, with preferably a similar or better measurement time and signal-to-noise-ratio, can provide a more accurate determination of the Zeeman shift.

Here a solution is proposed by measuring the Zeeman shift using a measurement which has the feasiblity of measuring the Zeeman shift with 0.1 kHz accuracy, sufficient for the aimed accuracy in the determined isotope shift. The measurement is based on applying two rf fields separated in time, which was proposed by Ramsey in 1950 [206] and nowadays is a common and very successful measurement technique. In order to perform a Ramsey-type measurement we first need to show that we can create a coherent superposition of the different magnetic substates of  ${}^{4}\text{He}^{*}$ .

Ramsey-spectroscopy has been used before (in 1958) to determine the  $g_J$  factor of <sup>4</sup>He<sup>\*</sup>, and this was in an atomic beam experiment [306], which is a completely different regime than we are considering here. Furthermore this technique has been used in a rubidium F = 2 BEC to measure the Zeeman shift [307], and is also closely related to investigation of the dynamics of spinor BECs with alkali atoms [308–310]. Although we are interested in employing this technique to measure the Zeeman shift,

within this field <sup>4</sup>He<sup>\*</sup> would also be an interesting candidate due to its extremely linear Zeeman shift and large loss channels compared to the alkali atoms generally used.

#### Rabi oscillations between magnetic substates of <sup>4</sup>He<sup>\*</sup>

The  $m_J = 0, \pm 1$  magnetic substates of <sup>4</sup>He<sup>\*</sup> have a linear Zeeman shift of  $m_J \times 2\mu_B \approx m_J \times 2.8$  MHz/G. For the  $m_J = 0$  state, the leading (second) order Zeeman shift is quadratic and calculated to be 2.275 mHz/G<sup>2</sup> [311, 312]. For the typical magnetic fields in our experiment (< 10 G) this is a completely negligible effect.

The S states have no hyperfine structure in  ${}^{4}\text{He}$ , as this isotope has no nuclear spin. In cases of fine and hyperfine structure (such as for the L > 0 states in <sup>4</sup>He or for <sup>3</sup>He, which has nuclear spin I = 1/2). the LS-coupling scheme breaks down at large Zeeman shifts that are comparable to the fine or hyperfine splitting. This is called the Paschen-Back regime [287] and adds a nonlinear energy shift to the magnetic sublevels, as has been shown for the P states in  ${}^{4}\text{He}$  [313, 314] and  ${}^{3}\text{He}$ [315, 316], and for the <sup>3</sup>He<sup>\*</sup> states although the effect is absent for the <sup>3</sup>He<sup>\*</sup> F = 3/2,  $m_F = \pm 3/2$  states that we generally work with [315, 317– 319]. For the 2S states in <sup>4</sup>He the Paschen-Back effect could only occur if the Zeeman splitting is comparable to the energy splitting to the next S state (~ 100 THz), occuring at fields of >  $10^7$  G [320, 321]. The  $2 {}^{1}S_{0}$  state has a second order Zeeman shift of 3.193 mHz/G<sup>2</sup> [311, 312], and no relevant Paschen-Back effect as there is only one hyperfine state. Relativistic and higher-order QED corrections to the q factor of all states start playing a role at the  $10^{-5}$  level (i.e. ~ 10 Hz for our typical Zeeman shifts of  $\sim 1$  MHz) and are currently negligible [322, 323].

For the magnetic fields that we operate at, the magnetic sublevels of  ${}^{4}\text{He}^{*}$  are a highly symmetric three-level system. This means that the levels form a cascaded level system where multiple levels couple symmetrically when applying a single rf frequency.

Denoting the  $m_J = 0, \pm 1$  states with state vectors  $|0\rangle$ , and  $|\pm\rangle$ , the total time-dependent state vector  $|\phi(t)\rangle$  for a <sup>4</sup>He<sup>\*</sup> atom is

$$|\phi(t)\rangle = \sum_{i=0,+,-} C_i(t)|i\rangle.$$
 (5.12)

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When applying an oscillating magnetic field  $\vec{B}_{\rm rf}(t)$  the Hamiltonian H of this three-level system is

$$H = g_J \mu_B \vec{S} \cdot \vec{B}(t) = \hbar \begin{pmatrix} \omega_0 & \frac{\Omega}{\sqrt{2}} e^{-i\omega t} & 0\\ \frac{\Omega}{\sqrt{2}} e^{i\omega t} & 0 & \frac{\Omega}{\sqrt{2}} e^{-i\omega t}\\ 0 & \frac{\Omega}{\sqrt{2}} e^{i\omega t} & -\omega_0 \end{pmatrix}, \quad (5.13)$$

where  $\vec{B}(t) = \vec{B}_0 + \vec{B}_{\rm rf}(t)$  is the total magnetic field,  $\omega$  the rf frequency,  $\omega_0 = g_J \mu_B B_0 / \hbar$  the Larmor frequency (which can be similar to the Zeeman shift), and  $\Omega$  the effective Rabi frequency of the system defined as  $\Omega^2 = \Omega_R^2 + \delta^2$  with  $\Omega_R = g_J \mu_B |\vec{B}_{\rm rf}|/2\hbar$  the pure Rabi frequency and  $\delta = \omega - \omega_0$  the detuning from the transition.

The time-dependent Schrödinger equation  $H|\phi(t)\rangle = i\hbar \frac{\partial}{\partial t} |\phi(t)\rangle$  gives the three coupled differential equations that describe the populations of the substates:

$$i\frac{dC_{+}}{dt} = \omega_{0}C_{+}(t) + \frac{\Omega}{\sqrt{2}}\exp(-i\omega t)C_{0}(t),$$
  

$$i\frac{dC_{0}}{dt} = \frac{\Omega}{\sqrt{2}}\exp(i\omega t)C_{+}(t) + \frac{\Omega}{\sqrt{2}}\exp(-i\omega t)C_{-}(t),$$
  

$$i\frac{dC_{-}}{dt} = -\omega_{0}C_{-}(t) + \frac{\Omega}{\sqrt{2}}\exp(i\omega t)C_{0}(t).$$
  
(5.14)

In the case of zero detuning  $(\delta = 0, \omega = \omega_0)$ ,  $\Omega = \Omega_R$  and Eqs. 5.14 can be solved analytically. Starting with a spin-polarized atom in  $m_J = +1$ state (i.e.  $C_+(0) = 1, C_0(0) = C_-(0) = 0$ ), the populations evolve as

$$|C_{+}(t)|^{2} = \cos^{4}\left(\frac{\Omega_{R}t}{2}\right),$$
  

$$|C_{0}(t)|^{2} = \frac{1}{2}\sin^{2}(\Omega_{R}t),$$
  

$$|C_{-}(t)|^{2} = \sin^{4}\left(\frac{\Omega_{R}t}{2}\right).$$
  
(5.15)

This result is shown in Figure 5.4. It is thus possible to transfer 50% of the atoms to  $m_J = 0$ , while 100% can be transferred to  $m_J = -1$ .

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**Figure 5.4** – Normalized population evolution of the  $m_J = +1$  (blue, full),  $m_J = 0$  (black, dotted), and  $m_J = -1$  (red, dashed) states of <sup>4</sup>He<sup>\*</sup> according to Equations 5.14. We see that a  $\pi/2$  pulse transfers 50% of the atom to the  $m_J = 0$  state and a  $\pi$  pulse transfer all atoms to  $m_J = -1$ .

In the experiment we prepared  $10^6$  atoms, either slightly above the transition to BEC or primarily in the BEC, in the  $m_J = +1$  state at an earthmagnetic field strength of about 0.5 G. An rf pulse was then applied for a varying time and the population of the three magnetic substates was measured by absorption imaging after 8 ms expansion in a magnetic field gradient. Figure 5.5 shows the experimental results for Rabi oscillations in case of a BEC, which provides the best contrast in absorption imaging. As it is difficult to normalize the different pictures we have normalized to the total number of atoms at each individual measurement time. Poisson noise in the atom number then introduces an up to 10% error in the normalized population of the different  $m_J$  states.

Figure 5.5 clearly shows that Eqs. 5.14, with a Rabi frequency  $\Omega_R = 2\pi \times 23.348(3)$  kHz, represent the measurements very well in general. The signal to noise ratio stays the same, but a change in the Rabi frequency is apparent for long Rabi pulse lengths. This is possibly caused by transient behavior of the rf amplifier system, but currently not



**Figure 5.5** – Relative populations of the  $m_J = +1$  (blue squares),  $m_J = 0$  (black dots), and  $m_J = -1$  (red diamonds) states of <sup>4</sup>He<sup>\*</sup> as function of rf pulse length, for atoms starting in the spin-polarized  $m_J = +1$  state at t = 0. The error bars are largest near a population of 50% and zero at 0% and 100% as a consequence of normalization.

limiting for creating  $\pi/2$  and  $\pi$  pulses for which we use the shortest Rabi pulse length available (~ 10  $\mu$ s and ~ 20  $\mu$ s, respectively). A  $\pi$  pulse transfers all atoms to the  $m_J = -1$  state while for a  $\pi/2$  pulse we find equal numbers of atoms in each of the spin-stretched  $m_J = \pm 1$  states as expected. However, there is a clear deficit of atoms in the  $m_J = 0$ state; where 50% is expected only 25% is observed. This is illustrated in Figure 5.6 (left), which shows an image of the three magnetic substates after a  $\pi/2$  pulse on a BEC and 8 ms expansion time. We attribute this deficit of  $m_J = 0$  atoms to Penning ionization within the expanding  $m_J = 0$  cloud. Being strongly dependent on density [132] we tested this by preparing a thermal cloud with about a factor 10 lower density. Indeed we see in Figure 5.6 (right) that after the same expansion time now the number of  $m_J = 0$  atoms in the  $m_J = \pm 1$  states, and of the order of 50%.



**Figure 5.6** – False-color absorption images taken after a  $\pi/2$  pulse in case of a Bose-Einstein condensate (left) and a thermal cloud (right). The pictures and integrated column density below it are taken 8 ms after release from the dipole trap and expansion in a magnetic field gradient. The thermal cloud expands much faster and thereby diminishes Penning ionization losses. The expected relative populations of the three magnetic substates are thus recovered.

#### Ramsey oscillations in <sup>4</sup>He<sup>\*</sup>

The Rabi oscillations between the magnetic substates can be used to apply  $\pi/2$  and  $\pi$  pulses in a controlled way, and the next step is to implement a two-pulse configuration which will allow observation of the temporal evolution of the magnetic substates. After creating a superposition of states using the Rabi pulse as described in the previous section, the eigenstates  $|+\rangle$ ,  $|0\rangle$ , and  $|-\rangle$  freely evolve with a phase factor  $\exp(-iEt/\hbar)$ , where E is the total energy of the state. If the Zeeman shift is the only relevant energy we can substitute  $E_{\pm} = \pm \hbar\omega_0$  for the  $|\pm\rangle$  states, and  $E_0 = 0$  for the  $|0\rangle$  state. Thus the magnetic substates can freely precess and acquire a phase for a certain time until a second Rabi pulse (identical to the first one) is applied. The resulting projection of states will then oscillate as function of the acquired phase, from which we can extract the Larmor frequency  $\omega_0$  and thus the Zeeman shift.

In a more mathematical description the Rabi pulses have a duration  $\tau$ and the states freely precess for a time  $\Delta T$ , and this process is described using the unitary operators  $U_{\text{Rabi}}(\tau)$  and  $U_{\text{free}}(\Delta T)$  to calculate the final state vector as

$$|\phi(t+t_{\rm seq})\rangle = U_{\rm Rabi}(\tau)U_{\rm free}(\Delta T)U_{\rm Rabi}(\tau)|\phi(t)\rangle, \qquad (5.16)$$

where  $t_{\text{seq}} = 2\tau + \Delta T$ . In this notation we assume that the initial phases of the Rabi pulses are the same for all free precession times  $\Delta T$ . If one were to correct for this phase the final populations would oscillate with the detuning  $\delta$  of the rf frequency with respect to the Larmor frequency instead of the pure Larmor frequency. To measure the population of the different magnetic substates with good signal-to-noise we choose  $\tau = \pi/(2\Omega_R)$  to create a  $\pi/2$  pulse. In this case the analytic form of the time evolution operator is

$$U_{\text{Rabi}}\left(\frac{\pi}{2\Omega_R}\right) = \begin{pmatrix} \frac{1}{2} & -\frac{i}{\sqrt{2}} & -\frac{1}{2} \\ -\frac{i}{\sqrt{2}} & 0 & -\frac{i}{\sqrt{2}} \\ -\frac{1}{2} & -\frac{i}{\sqrt{2}} & \frac{1}{2} \end{pmatrix}.$$
 (5.17)

From Eq. 5.16 and using the exact form for a  $\pi/2$  pulse sequence, an analytical expression for the magnetic sublevel populations after the Ramsey measurement can be calculated:

$$|C_{+}(\Delta T)|^{2} = \sin^{4}\left(\frac{\omega\Delta T}{2}\right),$$
  

$$|C_{0}(\Delta T)|^{2} = \frac{1}{2}\sin^{2}(\omega\Delta T),$$
  

$$|C_{-}(\Delta T)|^{2} = \cos^{4}\left(\frac{\omega\Delta T}{2}\right),$$
  
(5.18)

which looks very similar to the result of a single Rabi pulse.

This time-dependent behavior is also measured in the laboratory. The  $\pi/2$  pulse is generated using a Agilent 33220A arbitrary waveform generator which produces a burst of N cycles of rf with well-defined amplitude, phase, and frequency, and the timing between the pulses was initially triggered by the TTL pulses from the overall experimental control system, but has been upgraded to a two-pulse sequence from a second Agilent arbitrary waveform generator to reduce timing jitter. Using the same method to observe the populations of the magnetic substates as described in the previous section whilst scanning  $\Delta T$ , we observe oscillations with a frequency given by the Larmor frequency of ~ 1.5 MHz  $(1/f \approx 0.7 \ \mu s)$  as is shown in Figure 5.7.

Due to Penning ionization losses in the  $m_J = 0$  state, we cannot reconstruct the full state vector  $|\phi(t + t_{seq})\rangle$  as the populations are no longer conserved. Therefore we can calculate a normalized  $m_J = +1$  population  $|C_+(t + t_{seq})|^2_{\text{norm}}$  which is purely based on the populations of the  $m_J = \pm 1$  states as

$$|C_{+}(t+t_{\rm seq})|_{\rm norm}^{2} = \frac{|C_{+}(t+t_{\rm seq})|^{2}}{|C_{+}(t+t_{\rm seq})|^{2} + |C_{-}(t+t_{\rm seq})|^{2}}.$$
 (5.19)

The advantage of this system is that, due to the symmetry between the  $m_J = \pm 1$  states, this provides a signal that is simple to interpret, but it can no longer provide information on the decoherence of the signal as we cannot reconstruct the norm of the total state vector  $|\phi(t + t_{\text{seq}})\rangle$ . From Figure 5.7 it is clear that at larger  $\Delta T$  the signal-to-noise becomes less and a form of dephasing starts to appear. The signal-to-noise cannot decrease due to Penning ionization as the 50  $\mu$ s timescale is too short for significant losses at these densities (which are  $10^{12}$ - $10^{13}$  cm<sup>-3</sup>), and at this timescale Penning ionization does not lead to decoherence. A possible source may be the experimental rf amplifier noise at the timescale of  $\Delta T$ . On the other hand, as the acquisition of a single measurement shot takes about 15 s we suspect the observed decoherence may also be due to slow drifts and shot-to-shot variations in the magnetic field strength.

#### Mean-field effects on the Zeeman shift

For the explanation of the Ramsey spectroscopy experiment, set up to measure the Zeeman shift, we have not yet considered the fact that





Figure 5.7 – Relative population of the  $m_J = +1$  state of  ${}^4He^*$  as function of  $\pi/2$ -pulse delay  $\Delta T$ , for atoms starting in the  $m_J = +1$ state at t = 0. Below an expanded part for short and long time delay showing that the oscillations remain visible up to  $45 \ \mu s$ . The red line is a fit to the signal using Eq. 5.19.

mean-field interactions between the different magnetic substates have an effect on the phase evolution of the individual substates. The meanfield effects are straightforward to implement in this description, as the free evolution of the magnetic substates is given by the phase factor  $\exp(-iEt/\hbar)$ , where we now substitute the mean-field energy to the eigenenergy of the states  $E_{\pm} = \mu_{\pm}$  and  $E_0 = \mu_0$ . As the mean-field energy is only relevant for high densities, we associate  $\mu$  with the chemical potential of a BEC.

We use the Gross-Pitaevskii (GP) description [202] of the three BECs that are present once we apply a  $\pi/2$  pulse. For a state *i* out of the three states *i*, *j*, *k*, the wavefunction  $\psi_i$  of the condensate is described by the GP equation

$$\left(-\frac{\hbar^2 \nabla^2}{2m} + V_{\text{int}} + N_i g_{i,i} |\psi_i|^2 + N_j g_{i,j} |\psi_j|^2 + N_k g_{i,k} |\psi_k|^2\right) \psi_i = \mu_i \psi_i,$$
(5.20)

where  $V_{\text{int}}$  is the internal energy of the state and  $N_i$  the atom number in state *i*. The interaction parameter  $g_{i,j}$  is defined as

$$g_{i,j} = \frac{4\pi\hbar^2 a_{i,j}}{m},$$
 (5.21)

where  $a_{i,j}$  is the *s*-wave scattering length for atoms in states *i* and *j*. From Ref. [143] we extract the real and imaginary scattering lengths (where the latter are related to loss channels) as shown in Table 5.4. The imaginary scattering length, related to the loss channels, is divided into an exothermic channel and a degenerate channel, where the exothermic channel is associated with Penning ionization and the degenerate channel to collisions which change the magnetic substates but conserves energy (e.g. a  $m_J = +1, m'_J = -1$ )  $\rightarrow (m_J = 0, m'_J = 0)$  collision). Further details are discussed in Appendix 5.A, but note that the scattering lengths of the  $(m_J, m'_J) = (1, 1), (1, 0), (-1, 0), \text{ and } (-1, -1)$  are all the same, as all of these collisions only take place in the quintet potential [143]. Of the scattering lengths mentioned in Table 5.4, the only measured scattering length is  $a_{1,1} = 142.0(1) a_0$  [152].

In the Thomas-Fermi approximation the kinetic term in the GP equation is neglected with respect to the other energies, and the problem simplifies **Table 5.4** – Real and imaginary scattering lengths for collisions between the various magnetic substates of <sup>4</sup>He<sup>\*</sup>, denoted as  $(m_J, m'_J)$ , where  $a^{im,ex}$  and  $a^{im,deg}$  refer to exothermic and degenerate loss channels, respectively (see Appendix 5.A). The total imaginary scattering length is defined as  $a^{im} = a^{im,ex} + k \times a^{im,deg}$  where  $k = \sqrt{2\mu E}/\hbar$  with  $E = \frac{3}{2}k_BT$  the average collision energy and  $\mu = m/2$  the reduced mass. For  $T \approx 0.2 \ \mu K$  we get  $k \approx 1.6 \times 10^6 \ m^{-1} = 8.3 \times 10^{-5} \ a_0^{-1}$ . The scattering lengths are taken from figures in Ref. [143].

Collision channel	$a^{\mathrm{re}}\left[a_{0}\right]$	$a^{\text{im,ex}} [a_0]$	$a^{\mathrm{im,deg}} \left[ a_0^2 \right]$	$a^{\mathrm{im}} [a_0]$
$(\pm 1, \pm 1), (\pm 1, 0)$	140	$4 \times 10^{-4}$	0	$4 \times 10^{-4}$
(0,0)	120	13	$3  imes 10^4$	15
(+1,-1)	60	26	$3  imes 10^4$	28

to the scalar expression

$$V_{\text{int}} + N_i g_{i,i} |\psi_i|^2 + N_j g_{i,j} |\psi_j|^2 + N_k g_{i,k} |\psi_k|^2 = \mu_i$$
(5.22)

In this limit the direct relationship between the wavefunction of the condensate and the density distribution  $n_i(\vec{r})$  is  $\psi_i = \sqrt{n_i(\vec{r})/N_i}$ , which gives

$$\mu_{+1} = V_{\text{int}}^{+1} + g_{+1,+1}(n_{+1} + n_0) + n_{-1}g_{+1,-1},$$
  

$$\mu_0 = V_{\text{int}}^0 + g_{+1,+1}(n_{+1} + n_{-1}) + n_0g_{0,0},$$
  

$$\mu_{-1} = V_{\text{int}}^{-1} + g_{+1,+1}(n_{-1} + n_0) + n_{+1}g_{+1,-1},$$
  
(5.23)

where the explicit spatial dependence is omitted for convenience and the equations are simplified using the fact that the scattering lengths (and therefore the interaction parameters) are equal except for the (0,0) and (+1,-1) collisions. The internal energies all contain the same ac Stark shift from the optical dipole trap as this is a scalar shift. This can be included but only results in a common phase evolution and will drop out in the final calculations; it is therefore neglected here. The Zeeman shift is included as a linear shift, defining the  $m_J = 0$  state as reference energy:  $V_{\text{int}}^0 = 0$  and  $V_{\text{int}}^{\pm 1} = \pm \hbar \omega_0$ , where  $\omega_0$  is the Larmor frequency.

Without interactions the energies  $V_{\text{int}}$  are the only relevant energies to describe the free evolution operator  $U_{\text{free}}$  as described in the previous section, but the full chemical potential of each state needs to be used to include the mean-field interactions. As long as the timescale of interest is shorter than the Penning ionization timescale, the chemical potentials fully describe the phase evolution of the three components of the BEC after a Rabi pulse.

When applying a perfect  $\pi/2$  pulse we get  $n_{+1} = n_{-1} = n/4$  and  $n_0 = n/2$ , where n is the initial density of the BEC. In this case

$$\mu_{+1} = \hbar\omega_0 + \frac{n}{4}(3g_{+1,+1} + g_{+1,-1}),$$
  

$$\mu_0 = \frac{n}{2}(g_{+1,+1} + g_{0,0}),$$
  

$$\mu_{-1} = -\hbar\omega_0 + \frac{n}{4}(3g_{+1,+1} + g_{+1,-1}).$$
  
(5.24)

To get an order of magnitude estimate of the mean-field effects on the total evolution, we calculate the mean-field energies  $E_{\rm mf}^{m_J}$  for the  $m_J$  states at a typical BEC density of  $10^{13}$  cm<sup>-3</sup>. We get

$$E_{\rm mf}^{\pm 1} = \frac{n}{4} (3g_{\pm 1,\pm 1} + g_{\pm 1,-1}) \approx h \times 2.0 \text{ kHz}, \tag{5.25}$$

$$E_{\rm mf}^0 = \frac{n}{2}(g_{\pm 1,\pm 1} + g_{0,0}) \approx h \times 2.2 \text{ kHz}, \qquad (5.26)$$

which are typically ~ 0.1% of the Zeeman shift experienced by the states. This is problematic when trying to measure the Zeeman shift beyond the kHz level. However, as the phase of each state evolves as  $\exp(-i\mu t/\hbar)$ , we should consider the differences between the chemical potentials of the states to see how the differential phase develops between the states:

$$\mu_{+1} - \mu_{-1} = 2\hbar\omega_0,$$

$$\mu_{+1} - \mu_0 = \hbar\omega_0 + \frac{n}{4}(g_{+1,+1} + g_{+1,-1} - 2g_{0,0}) \approx \hbar\omega_0 + \frac{n}{14}g_{+1,-1}.$$
(5.28)

We see that the mean field energy drops out as common mode when considering the phase evolution between the  $m_J = +1$  and  $m_J = -1$  states if the densities of both components are equal. Furthermore, the phase difference evolves with exactly twice the Larmor frequency by virtue of the extreme linearity of the Zeeman shift. In terms of sensitivity we can calculate the resulting energy shift  $\Delta E_{\rm mf}$  from a fractional density difference  $\Delta n/n$  between the two states at the typical experimental density of  $10^{13}$  cm<sup>-3</sup>:

$$\Delta E_{\rm mf} = \frac{\Delta n}{n} \times h \times 1.3 \text{ kHz.}$$
(5.29)

Thus, if we can get the populations to be stable within a few percent the shift is < 0.1 kHz. Also, from a practical point of view this method only involves the  $m_J = \pm 1$  states, which are much better imaged than the  $m_J = 0$  state (see Figure 5.6). By measuring the evolution of  $m_J = +1$  versus the  $m_J = -1$  state, as is shown in Figure 5.7, we indeed recover an oscillation frequency which is twice the Larmor frequency that we determined with the former rf resonance method.

To conclude, it might be surprising to learn that the mean-field interactions have a significant effect on the Zeeman shift measurements. This means that there is a systematic effect on a systematic effect for the high-precision transition frequency measurement that should be controlled sufficiently. By virtue of the linearity of the Zeeman shift and the symmetry of the collision channels (and therefore *s*-wave scattering lengths) between the different states, a Ramsey-type method of measurement is proposed which has the possibility of fully eliminating the mean-field effects from the Zeeman shift measurement. This method has currently not been fully implemented to substitute the former rf resonance method as used in [31, 35], but measurements as shown in Figure 5.6 have already shown that it is possible to reproduce equal populations in the  $m_J = +1$  and  $m_J = -1$  states with  $\Delta n/n \approx 10\%$ , which is already sufficient and can only be further improved by improving the stability of the rf amplifier.

## 5.4 Spectroscopy in the ${}^{4}\text{He}{}^{*}$ m<sub>J</sub> = 0 state

In Section 5.3 it is shown how 50% of the atoms of the spin-polarized  $m_J = +1$  BEC can be transferred to the  $m_J = 0$  state using a  $\pi/2$  pulse. Applying a magnetic field gradient after the  $\pi/2$  pulse whilst keeping the dipole trap on pushes the  $m_J = \pm 1$  state atoms out of the dipole trap, leaving a pure  $m_J = 0$  cloud of quantum degenerate <sup>4</sup>He<sup>\*</sup>. An

almost 100% transfer efficiency can be achieved using two Raman laser pulses at the 2  ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{0}$  transition [169] at the cost of an additional laser system. Apart from the factor of two in the atom number, the rf transfer method is easier to implement and maintain for our purposes. With the availability of a pure  $m_{J} = 0$  gas, we can explore a few new applications of  ${}^{4}\text{He}^{*}$ . As the  ${}^{4}\text{He}^{*}$  state has a second order Zeeman shift of only 2.275 mHz/G<sup>2</sup> [311, 312], it is much more insensitive to the magnetic field than other atoms, which has interesting applications for atom interferometry with  ${}^{4}\text{He}^{*}$  [90]. Another interesting application is for high-precision frequency metrology, as the differential second order Zeeman shift on the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$  transition is only 0.918 mHz/G<sup>2</sup> [311, 312]. Thus a direct measurement of the 2  ${}^{3}S_{1}(m_{J} = 0) \rightarrow 2 {}^{1}S_{0}$ transition is essentially Zeeman shift free. The possibility for measuring this transition in  ${}^{4}\text{He}$  would be very interesting as it directly eliminates a systematic effect from the error budget.

There are a few problems with working with ultracold <sup>4</sup>He<sup>\*</sup> in the  $m_J = 0$  state, as this state is not stable against Penning ionization in contrast to the spin-stretched  $m_J = \pm 1$  states. This is the main reason why spectroscopy in a ultracold cloud of pure  $m_J = 0$  atoms was never thought to be feasible.

#### Lifetime of a $m_J = 0$ gas

As an  $m_J = 0$  gas is not stable against Penning ionization, the lifetime of the gas in the trap should be estimated to examine the feasibility of performing spectroscopy. Measurements of the lifetime of the  $m_J = 0$ BEC already show that it fully annihilates within a few ms, leaving only a thermal gas of  $m_J = 0$  atoms. Therefore I will focus here on the feasibility of measurements in a thermal gas of  $m_J = 0$  atoms. The general rate equation (derived in Appendix 5.B) is:

 $\frac{\partial N}{\partial t} = -\Gamma N - L_2 \frac{N^2}{V_{\text{eff}}} - L_3 \frac{8}{\sqrt{27}} \frac{N^3}{V_{\text{eff}}^2},\tag{5.30}$ 

where  $\Gamma$  represents the loss rate due to background losses,  $L_2$  and  $L_3$  are loss rate constants related to two- and three-body collisions (for previous work concerning  $L_3$  in our group see Refs. [156, 324, 325]), and  $V_{\text{eff}}$  is an effective volume of the cloud. For this discussion the  $L_3$  term can be neglected as the density of the thermal gas is already sufficiently low. We typically have  $1/\Gamma \approx 10$  s which is much longer than the lifetime due to Penning ionization and can therefore also be neglected for the purpose of these calculations. Within these approximations the loss rate equation is

$$\frac{\partial N}{\partial t} = -L_2^{00} \frac{N^2(t)}{V_{\text{eff}}},\tag{5.31}$$

for our pure  $m_J = 0$  gas, where N is the number of  $m_J = 0$  atoms and  $L_2^{00}$  the loss rate related to (0,0) collisions. From theory [143],  $L_2^{00} = K_{00}^{inel}$  and the scattering lengths as given in Table 5.4 can be used to calculate the thermally averaged loss rate  $\langle K_{00}^{inel} \rangle$  as discussed in Appendix 5.A. This calculation can be compared to experimental data from the Palaiseau group [155], as they have measured the spindependent loss rates of an ultracold metastable helium mixture in an optical dipole trap. Their result, for a thermal gas at a temperature of 2.0  $\mu$ K, is  $L_2^{00} = 6.6(4)_{\text{stat}}(1.7)_{\text{syst}} \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$  and shown together with the calculations in Figure 5.8. Note that this specific case, a spinpolarized cloud of  $m_J = 0$  atoms, has not been calculated before but the data and theory seem to be in good agreement with each other. For an unpolarized thermal cloud the loss rate coefficient  $K_{\text{unpol}}$  was measured before [149] and is related to  $K_{00}^{inel}$  as  $K_{\text{unpol}} = \frac{1}{6}K_{00}^{inel}$ , which is in agreement with the result by Partridge and coworkers (see Figure 10 in Ref. [132]).

Figure 5.8 shows that for temperatures below 1  $\mu$ K the loss rate converges to a constant rate, and the calculated loss rate and the experimental result [155] can be assumed to be good estimates for the expected loss rates. The inelastic loss rate is  $\langle K_{00}^{\rm inel} \rangle \approx 2.5 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$ , and agrees with the observation that the BEC (with a peak density of  $10^{13} \text{ cm}^{-3}$ ) is lost within a few ms. For the thermal fraction with a peak density which is ten times lower, the lifetime would be 10's of ms. This may be too short for performing spectroscopy.

Surprisingly, a measurement of the lifetime of the thermal  $m_J = 0$  cloud in our experiment reveals a two-body loss rate that is almost an order of magnitude lower than expected. This means that still a significant number of atoms is present after even 0.5 s. Although calibration of the MCP detector could play a role here, a more probable possibility is that



**Figure 5.8** – The (thermally averaged) loss rate coefficient  $\langle K_{00}^{inel} \rangle$  calculated for a wide range of temperatures. The shaded area indicates a 40% uncertainty in the theoretical calculations [143]. The datapoint is the experimental determination by Partridge et al. [155].

these atoms reside in the 'arms' of the optical dipole trap as the density is low enough in the arms to have low collision rates. Absorption images of the  $m_J = 0$  clouds (see Figure 5.6) show a small signal-to-noise ratio due to the low densities, but they seem to confirm this. Nonetheless, from the time-of-flight measurements on the MCP detector a good fit of a Maxwell-Boltzmann distribution is still obtained, indicating that the atoms are thermalized.

The low atom number is not a limiting factor as the gain of the MCP detector can be increased. For an absolute atom number determination the MCP should then be recalibrated, but as we measure the transition as a relative loss signal, the absolute signal is not relevant.

To conclude, Penning ionization losses are severe and definitely limiting the allowable interaction time to do spectroscopy in our experiment. However, at very low densities the gas has sufficient lifetime to do the experiment whilst atoms can still be detected by increasing the gain of the MCP detector.

#### **Transition** rates

As the lifetime of the  $m_J = 0$  cloud is short, the excitation rate of the transition should be calculated to estimate if there is sufficient signal. The 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$  transition rate can be different when starting from the  $m_J = +1$  state or starting from the  $m_J = 0$  state as the former requires a  $\sigma^{-}$  transition and the latter a  $\pi$  transition. The Clebsch-Gordan coefficients for both transitions are exactly the same, so the transition can in principle be made using the same spectroscopy laser power. But the actual experiment geometry should also be taken into account.

The current setup of the spectroscopy beam is optimized to have a linear polarization orthogonal to the quantization axis of the atoms, such that the polarization vector can be decomposed in equal right-hand and lefthand circular polarization components, of which one will induce the  $\sigma^{-}$ transition (as discussed in more detail in Section 4.B). Apart from small misalignment of the spectroscopy laser beam with respect to the quantization axis or impurity of the polarization state of the spectroscopy laser, there is no projection of the polarization vector along the quantization axis, which is necessary to induce a  $\pi$  transition. By rotating the polarization by 90°, the linear polarization will project on the quantization axis with a small angle of  $9.5^{\circ}$  (half the dipole trap beam crossing angle of 19°). As the excitation rate scales with the square of the electric field, the effective projection will be  $\sin(9.5^{\circ})^2 \approx 3\%$  of the total available power. Compared to the original configuration, which has a 50% efficiency, this is a reduction in excitation rate for the  $\pi$  transition from the  $m_J = 0$  state by a factor of 20. The excitation rate could be improved by 5% to 200% of the current excitation rate by changing the spectroscopy beam geometry (or the quantization axis of the atoms) in the current experiment. This is beyond the current goal of simply observing the transition.



**Figure 5.9** – First direct observation of the magnetic field insensitive  $2 {}^{3}S_{1} (m_{J} = 0) \rightarrow 2 {}^{1}S_{0}$  transition for a thermal cloud of atoms with  $T \approx 4 \ \mu K$ . A Voigt line shape fit (red line) with a Lorentzian FHWM of 4.5 kHz reveals a Gaussian FHWM of 47(1) kHz, in agreement with the Doppler width of 46 kHz at this temperature. The frequency axis is offset with respect to the centroid frequency  $f_{0,\text{fit}}$  determined from the fit.

### Observing the 2 ${}^{3}S_{1}$ $(m_{J} = 0) \rightarrow 2 {}^{1}S_{0}$ transition

Direct observation of the 2  ${}^{3}S_{1}$   $(m_{J} = 0) \rightarrow 2 {}^{1}S_{0}$  transition in a thermal cloud of  ${}^{4}\text{He}^{*}$  at  $T \approx 4 \ \mu\text{K}$  is shown in Figure 5.9. The on-resonance loss of ~ 40% of the atoms is achieved after allowing a spectroscopy beam interaction time of 4 s, which is large compared to the lifetime of the gas. To have a signal at all we increased the gain of the MCP by approximately a factor 4. Even with the large losses, the signal has a good signal-to-noise ratio and can be repeatedly observed.

Obviously the linewidth of the transition is much broader than for a BEC, which can be measured from  $m_J = +1$ . Using a Voigt line shape and fixing the Lorentzian contribution to the lineshape to the spectroscopy laser linewidth of 4.5 kHz as determined in the line shape work

in Chapter 4, the determined Gaussian FWHM is 47(1) kHz. This is in agreement with the calculated Doppler width of ~ 46 kHz based on a temperature of  $T \approx 4 \ \mu \text{K}$  determined from the time-of-flight measurements.

The clear and direct observation of the  $m_J = 0$  transition is exciting as it shows that it is feasible to perform a practically Zeeman shift-free measurement of the 2  ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$  transition in <sup>4</sup>He. However, there are two (related) concerns that need to be fully understood before this transition can be used for the high-precision measurements. The first observation is that the ac Stark shift at which the transition is observed. is approximately (not exactly) half the ac Stark shift of the  $m_J = +1$ transition. This difference can be explained by the earlier observation that the  $m_I = 0$  atoms predominantly reside in the 'arms' of the optical dipole trap where they experience only approximately half of the ac Stark shift compared to the atoms in the center of the trap (as is the case for spectroscopy in the BEC). Second, as was discussed in the previous section and in the previous argument, the atoms are residing in the arms of the ODT and the exact line shape model is not known. However, as the atoms appear to be thermalized a Voigt line shape fit seems the most appropriate, and the agreement with the determined Gaussian line width is an indicator that the width of the observed line is understood for a simple thermal gas model. For future frequency metrology the spatial and momentum distribution of the  $m_J = 0$  atoms in the optical dipole trap should be better understood, because it is possible that the atoms are not fully thermalized as the Penning ionization losses are so strong.

To conclude, using the coherent rf transfer of 50% of the  $m_J = +1$ BEC atoms to the  $m_J = 0$  state, we are able to create a pure cloud of  $m_J = 0$  atoms in the optical dipole trap. Due to Penning ionization losses we are only able to maintain the thermal atoms, as the  $m_J = 0$  BEC is destroyed within a few ms, and we are able to excite  $2 {}^{3}S_1 (m_J = 0) \rightarrow 2 {}^{1}S_0$  transition with sufficient signal-to-noise to perform spectroscopy. Although there are still some remaining issues to be solved, the prospect of using this transition for <sup>4</sup>He<sup>\*</sup> is very exciting in the context of significantly reducing the error budget.

# 5.5 Prospects for <sup>3</sup>He<sup>\*</sup>

The majority of this chapter deals with improving transition frequency measurements in  ${}^{4}\text{He}^{*}$ , but for the nuclear charge radius difference determination also the  ${}^{3}\text{He}^{*}$  measurements need to be improved. Several aspects will be discussed here that should be paid attention to in the future experiments.

#### Absence of collisions

Due to the absence of s-wave collisions in an ultracold degenerate Fermi gas of  ${}^{3}\text{He}^{*}$  atoms, the gas will not rethermalize during excitation. This means that atoms are not redistributed in the trap states during the excitation, and only atoms with excitation energies within the spectral width of the laser will be excited. This is a big contrast with the  ${}^{4}\text{He}^{*}$  measurement where it is possible, for some detunings, to excite all atoms from the trap as they rethermalize very efficiently. Essentially this means that the signal (i.e. the depletion of atoms from the trap) is quickly saturated. The exact circumstances for performing spectroscopy (laser linewidth, transition linewidth) should therefore be carefully weighed against each other in order to optimize the signal-to-noise in the measurement.

#### Line shape and statistics

As discussed in Chapter 4, the <sup>3</sup>He<sup>\*</sup> line shape is very broad due to the Fermi-Dirac statistics governing the spatial and momentum distribution in the trap. With the implementation of a magic wavelength trap, the differential ac Stark shift will be reduced significantly, improving the accuracy on the ac Stark shift extrapolation. Another advantage is the reduction (ideally a removal) of the asymmetry in the <sup>3</sup>He<sup>\*</sup> line shape, as it is solely caused by the differential ac Stark shift.

Unfortunately the linewidth of the transition in <sup>3</sup>He<sup>\*</sup> cannot be simply reduced as it is essentially given by the Fermi energy of the gas. As the Fermi energy scales as  $E_F \propto N^{1/3}$  (see Eq. 2.19), reduction of the linewidth in a harmonic trap can only be achieved by reducing the number of atoms. At first sight this seems to reduce the signal-to-noise ratio, but we need to take the signal saturation effect as discussed in the previous section into account. When reducing the number of atoms in the trap the transition linewidth will reduce, and the laser linewidth will be able to excite a larger fraction of atoms from the trap, thereby improving the signal-to-noise of the measurement. The relative shot-toshot fluctuations in the initial atom number will increase for decreasing atom number as  $N^{-1/2}$ . With a laser linewidth  $\Gamma$  and a transition linewidth  $\gamma \propto E_F \propto N^{1/3}$ , we can estimate that the relative signal scales as  $\Gamma/\gamma$ , and the signal-to-noise scales as SNR  $\propto \sqrt{N}(\Gamma/\gamma) \propto N^{1/6}\Gamma$ .

From this back-of-the-envelope estimate we already learn that we cannot improve the signal-to-noise by reducing the number of atoms. However, from a spectroscopy point-of-view an additional payoff lies in the improved spectral resolution of the line, which is linearly related to the linewidth  $\gamma$  of the transition. As the spectral resolution scales as  $\gamma \propto N^{1/3}$ , we see that we can win in spectral resolution faster than we lose signal-to-noise in the atom numbers and this is a route worth exploring. Ofcourse the spectral resolution cannot exceed the laser linewidth of 4.5 kHz, but this coincides with less than 100 fermions in the trap which would be a difficult feat to accomplish anyway.

#### Zeeman shift

The hyperfine metastable state in which we do spectroscopy in <sup>3</sup>He<sup>\*</sup> has F = 3/2, and thus no  $m_F = 0$  state available. This means that the Zeeman shift cannot be eliminated in the same way as for <sup>4</sup>He<sup>\*</sup> and it simply has to be measured as accurately as possible.

The Zeeman shift of the spin-stretched <sup>3</sup>He<sup>\*</sup> ( $F = 3/2, m_F = \pm 3/2$ ) states is<sup>2</sup>  $\pm (g_J + g_I/2)\mu_B B$ , and for the 2 <sup>1</sup>S<sub>0</sub> ( $F = 1/2, m_F = \pm 1/2$ ) states it is  $\pm (g_I/2)\mu_B B$ . None of these states show nonlinear behavior due to the Paschen-Back effect or hyperfine interactions [315, 317–319], which makes the system simple to describe. The differential Zeeman shift on either 2 <sup>3</sup>S<sub>1</sub> ( $F = 3/2, m_F = \pm 3/2$ )  $\rightarrow 2$  <sup>1</sup>S<sub>0</sub>( $F = 1/2, m_F = \pm 1/2$ )  $\sigma^+$  or  $\sigma^-$  transition is  $\pm g_J \mu_B B$  and therefore identical to the Zeeman shift measured using the proposed Ramsey spectroscopy method in <sup>4</sup>He<sup>\*</sup>. This method certainly has the possibility to achieve sub-kHz ac-

<sup>&</sup>lt;sup>2</sup>Here  $g_J = 2.00231930436182(52), g_I = 2.317481916(28) \times 10^{-3}$ , and  $\mu_B = 927.4009994(57)$  J/T [82].

curacy and is therefore an appropriate candidate for the final metrology experiment.

Another option is to acquire the aforementioned  $\sigma^+$  and  $\sigma^-$  transitions in a staggered way. By this we mean that while scanning over the transition, each measurement alternates between the two transitions which are detuned by  $\sim 3$  MHz from each other. With the current spectroscopy beam geometry and polarization this poses no problem as both transitions can be excited, and with the tunable offset frequency in the transfer lock of the spectroscopy laser (i.e. the direct digital synthesizer, DDS) the laser frequency can be changed between two measurements without any problems. Furthermore, the control software of the experiment already allows such type of measurement as it is currently used to alternate between measurements with and without spectroscopy light in order to properly normalize the signal. The advantage of this method is that the two transitions are acquired in the same period of time, minimizing any effects of magnetic field drift. Such drifts can be observed using the Ramsey method, but still depend on interpolation between separate measurements.

# 5.A Appendix: Collisions between two <sup>4</sup>He<sup>\*</sup> $m_J = 0$ atoms

The s-wave scattering length a of a collision between two atoms in specific states leads to an elastic scattering cross section  $\sigma = 4\pi a^2$  in the ultracold collision regime. However, things get more complicated when including loss processes such as Penning ionization, as this is an inelastic scattering process. Furthermore, the total collision process typically has to include multiple molecular potentials due the different possible scattering processes.

Penning ionization (PI) and associative ionization (AI) in <sup>4</sup>He<sup>\*</sup> occur in the triplet ( $\vec{S} = 1$ ) or singlet ( $\vec{S} = 0$ ) potentials. Any initial molecular potential which has  $\vec{S} = 0$  or  $\vec{S} = 1$  character at short internuclear distance will lead to strong losses due to the resulting ionization processes. In the well-known collision of two  $m_J = +1$  state atoms - which we denote as a (1,1) state - the total spin of the system is  $\vec{S} = 2$ , and orbital angular momentum  $\vec{L} = 0$ , leading to a pure  ${}^{5}\Sigma_{a}^{+}$  (quintet) potential. As this potential is unable to couple to the singlet and triplet potentials (except through weak spin-dipole processes [143]), it suppresses the PI and AI losses that would prevent achieving an ultracold gas of metastable helium atoms.

The story is different when two  ${}^{4}\text{He}^{*}$   $m_{J} = 0$  atoms collide - denoted as a (0,0) collision (and similar for (+1,-1) collisions). This constitutes a system that does not consist of identical spin-stretched states and the correct eigenstates need to be calculated in order to identify the relevant molecular potentials. For *s*-wave scattering the system decomposes as

$$|(J_1, m_1)(J_2, m_2)\rangle = |(1, 0)(1, 0)\rangle = \sqrt{\frac{2}{3}}|J = 2\rangle + \sqrt{\frac{1}{3}}|J = 0\rangle, \quad (5.32)$$

indicating that we have both the singlet and quintet potential contributing in this collision process. Note that for a (1,1) collision the coefficient of the  $|J = 2\rangle$  component would simply be 1, which shows that this collision purely occurs in a quintet potential. To incorporate the loss process taking place when two atoms collide in the  ${}^{1}\Sigma_{g}^{+}$  potential, the theoretical framework is laid out to include a so-called 'complex optical potential', which is a potential that includes a loss channel in the imaginary plane [143]. By varying the amplitude of the loss channel (also called the autoionization width), one can play around with the effective losses.

Using the complex optical potential, different loss processes can be incorporated and modeled. As the complex optical potential finds itself on the imaginary plane, the potential also has a associated complex scattering amplitude and therefore a complex scattering length. In Ref. [143] Leo *et al.* connect the complex scattering length between atoms of states  $\alpha$  and  $\beta$  as  $a_{\alpha,\beta}^{\text{im}}$ . The complex scattering length can be further subdivided into two categories of scattering amplitudes: those related to exothermal processes such as PI and to scattering to so-called degenerate states (which is the  $(0,0) \leftrightarrow (+1,-1)$  scattering process). The complex scattering length is defined as

$$a_{\alpha,\beta}^{\rm im} = a_{\alpha,\beta}^{\rm im,ex} + k \times a_{\alpha,\beta}^{\rm im,deg}, \qquad (5.33)$$

where  $k = \sqrt{2\mu E}/\hbar$  is the collision wavevector,  $\mu = m/2$  the reduced mass and  $E = \frac{3}{2}k_BT$  the collision energy in a thermal gas. This complex

scattering length has a finite effect on the elastic scattering cross section, as the loss rate still allows partial elastic processes. The total elastic scattering cross-section  $\sigma_{\alpha,\beta}^{\rm el}$  therefore reads

$$\frac{\sigma_{\alpha,\beta}^{\text{el}}}{4\pi} = (a_{\alpha,\beta}^{\text{re}})^2 + (a_{\alpha,\beta}^{\text{im}})^2, \qquad (5.34)$$

and the total inelastic rate is defined as  $K_{\alpha,\beta}^{\text{inel}} = K_{\alpha,\beta}^{\text{ex}} + K_{\alpha,\beta}^{\text{deg}}$ , where [326]

$$K_{\alpha,\beta}^{\text{ex}} = \frac{v}{k} \times 4\pi a_{\alpha,\beta}^{\text{im,ex}} = \frac{4\pi\hbar}{\mu} a_{\alpha,\beta}^{\text{im,ex}}, \qquad (5.35)$$

$$K_{\alpha,\beta}^{\text{deg}} = v \times 4\pi a_{\alpha,\beta}^{\text{im,deg}} = \sqrt{\frac{32\pi^2 E}{M}} a_{\alpha,\beta}^{\text{im,deg}}.$$
 (5.36)

Here the relative collision velocity v is defined as  $v = \sqrt{2E/\mu}$ , with E the collision energy (again,  $k = \sqrt{2\mu E}/\hbar$ ,  $E = \frac{3}{2}k_BT$  in a thermal gas and  $\mu = m/2$ ). Both the elastic and inelastic collision rate go to a constant value for  $E \to 0$ . One can now simply extract the calculated scattering lengths from Ref. [143] and use them to calculate loss rates. To calculate the loss rates, we have to take the velocity distribution into account when we want to discuss our experimental results. The proper normalised energy distribution that is used (ignoring Bose-Einstein or Fermi-Dirac statistics), is the Maxwell-Boltzmann distribution  $P^{\rm MB}(E)$ , which is defined as [114]

$$P^{\rm MB}(E)dE = 2\sqrt{\frac{E}{\pi}} \left(k_B T\right)^{-\frac{3}{2}} \exp\left(-\frac{E}{k_B T}\right) dE.$$
 (5.37)

The thermal average of any parameter A(E) is now defined as

$$\langle A \rangle = \int_0^\infty A(E) P^{\rm MB}(E) dE.$$
 (5.38)

As an example, consider the thermal average of the inelastic collision rate  $K_{\alpha,\beta}^{\text{inel}}$ . We get  $\langle K_{\alpha,\beta}^{\text{inel}} \rangle = \langle K_{\alpha,\beta}^{\text{ex}} \rangle + \langle K_{\alpha,\beta}^{\text{deg}} \rangle$ . As  $K_{\alpha,\beta}^{\text{ex}}$  is independent of energy, the calculation is trivial. The degenerate inelastic collision rate, however, depends on  $\sqrt{E}$  (see Eq. 5.36). The thermal average is calculated as

$$\langle K_{\alpha,\beta}^{\text{deg}} \rangle = \sqrt{\frac{32\pi^2}{\mu}} a_{\alpha,\beta}^{\text{im,deg}} \int_0^\infty \sqrt{E} P^{\text{MB}}(E) dE.$$
(5.39)

The integral gives  $\sqrt{4k_BT/\pi}$ , resulting in

$$\langle K_{\alpha,\beta}^{\text{deg}} \rangle = 8 \sqrt{\frac{2\pi k_B T}{\mu}} a_{\alpha,\beta}^{\text{im,deg}}.$$
 (5.40)

For comparison, if one were to simply substitute  $E = \frac{3}{2}k_BT$  in Eq. 5.36, the difference would be

$$\frac{\langle K_{\alpha,\beta}^{\text{deg}} \rangle}{K_{\alpha,\beta}^{\text{deg}}(T)} = \sqrt{\frac{8}{3\pi}} \approx 0.92, \qquad (5.41)$$

which is a small correction for rough estimates but can be important for the final analysis.

## 5.B Appendix: Trap loss calculations

In general, trap loss is described using the number density n of a gas:

$$\frac{\partial n}{\partial t} = -\Gamma n - L_2 n^2 - L_3 n^3, \qquad (5.42)$$

where  $\Gamma = 1/\tau$ ,  $L_2$ , and  $L_3$  represent the 1/e lifetime and two- and three-body loss rates, respectively. Time-dependence of the densities has been left out explicitly for convenience.

This description concerns particle densities that are relevant in understanding the actual physics. In our experiments, however, we generally measure atom numbers, which means that we have to integrate over the densities:

$$\frac{\partial N}{\partial t} = -\Gamma \int n \ d^3 \mathbf{r} - L_2 \int n^2 d^3 \mathbf{r} - L_3 \int n^3 d^3 \mathbf{r}.$$
 (5.43)

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As  $N = \int n \ d^3 \mathbf{r}$ ,  $\langle n \rangle = (1/N) \int n^2 d^3 \mathbf{r}$  and  $\langle n^2 \rangle = (1/N) \int n^3 d^3 \mathbf{r}$ , we get

$$\frac{\partial N}{\partial t} = -\Gamma N - L_2 N \langle n \rangle - L_3 N \langle n^2 \rangle.$$
(5.44)

The 3D harmonic oscillator potential approximation gives - for a thermal gas - the well-known Gaussian density distribution

$$n(\mathbf{r}) = n_0 \exp\left(-\frac{x^2}{2\sigma_x^2}\right) \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \exp\left(-\frac{z^2}{2\sigma_z^2}\right), \qquad (5.45)$$

where  $\sigma_i = \omega_i^{-1} \sqrt{k_B T/m}$  and  $\omega_i$  the trapping frequency. The normalization condition  $N = \int n(\mathbf{r}) d^3 \mathbf{r}$  provides the peak density  $n_0 = N(2\pi k_B T/(m\bar{\omega}^2))^{-3/2}$ , where  $\bar{\omega} = (\omega_x \omega_y \omega_z)^{1/3}$  is the geometric average oscillator frequency. The average density is given as

$$\langle n \rangle = \frac{n_0}{\sqrt{8}} = \frac{N}{V_{\text{eff}}},\tag{5.46}$$

where the effective volume is  $V_{\text{eff}} = (4\pi k_B T / (m\bar{\omega}^2))^{3/2}$ . For completeness the second moment of the density distribution is

$$\langle n^2 \rangle = \frac{n_0^2}{\sqrt{27}} = \frac{8}{\sqrt{27}} \left(\frac{N}{V_{\text{eff}}}\right)^2,\tag{5.47}$$

and the loss rate in terms of experimentally measurable quantities is

$$\frac{\partial N}{\partial t} = -\Gamma N - L_2 \frac{N^2}{V_{\text{eff}}} - L_3 \frac{8}{\sqrt{27}} \frac{N^3}{V_{\text{eff}}^2},$$
 (5.48)

as used in the main text.

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# List of Publications

The chapters in this thesis are based on the following publications:

- **Chapter 3:** R.P.M.J.W. Notermans and W. Vassen, "High-precision spectroscopy of the forbidden  $2 {}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$  transition in quantum degenerate metastable helium," *Phys. Rev. Lett.* **112**, 253002, 2014.
- Chapter 4: R.P.M.J.W. Notermans, R.J. Rengelink, and W. Vassen, "Comparison of spectral linewidths for quantum degenerate bosons and fermions," *Phys. Rev. Lett.* **117**, 213001, 2016.
- **Chapter 5:** R.P.M.J.W. Notermans, R.J. Rengelink, K.A.H. van Leeuwen, and W. Vassen, "Magic wavelengths for the  $2^{3}S \rightarrow 2^{1}S$  transition in helium," *Phys. Rev. A* **90**, 052508, 2014.
- Chapter 5: W. Vassen, R.P.M.J.W. Notermans, R.J. Rengelink, and R.F.H.J. van der Beek, "Ultracold metastable helium: Ramsey fringes and atom interferometry," *Appl. Phys. B* 122, 289, 2016.

### The author also contributed to the following publications:

(i) R.J. Rengelink, R.P.M.J.W. Notermans, and W. Vassen, "A simple 2 W continuous-wave laser system for trapping ultracold metastable helium atoms at the 319.8 nm magic wavelength," *Appl. Phys. B* 122, 122, 2016.

- (ii) S.H.W. Wouters, G. ten Haaf, R.P.M.J.W. Notermans, N. Debernardi, P.H.A. Mutsaers, O.J. Luiten, and E.J.D. Vredenbregt, "Performance predictions for a laser-intensified thermal beam for use in high-resolution focused-ion-beam instruments," *Phys. Rev.* A 90, 063817, 2014.
- (iii) D.E. Laban, A.J. Palmer, W.C. Wallace, N.S. Gaffney, R.P.M.J.W. Notermans, T.T.J. Clevis, M.G. Pullen, D. Jiang, H.M. Quiney, I.V. Litvinyuk, D. Kielpinski, and R.T. Sang, "Extreme ultraviolet interferometer using high-order harmonic generation from successive sources," *Phys. Rev. Lett.* **109**, 263902, 2012.
- (iv) T. Meijer, J.P. Beardmore, C.G.C.H.M. Fabrie, J.P. van Lieshout, R.P.M.J.W. Notermans, R.T. Sang, E.J.D. Vredenbregt, and K.A.H. van Leeuwen, "Structure formation in atom lithography using geometric collimation," *Appl. Phys. B* 105, 703–713, 2011.

## Summary

The helium atom provides an excellent testing ground for fundamental theory. It is a system that is initially simple enough to understand, vet the two electrons of a helium atom provide more complexity for calculations compared to the hydrogen atom as it is a three-body system compared to a two-body system. Moreover, there are additional electron-electron interactions. The most accurate theoretical framework to date, quantum electrodynamics, can predict the energies of the different lower electronic states of the helium atom with 9 to 10 digits of precision (for hydrogen it is more like 11–12 digits). By comparing these predictions to experimental determinations, the theory can be tested and in some cases provide more information about atomic properties, such as nuclear sizes, which are very difficult to measure using other methods. From an experimental perspective the helium atom is very interesting. In its ground state the stable electronic configuration makes it very difficult to let it interact with anything; it is a noble gas. However, in an excited state the atom becomes highly reactive. In the first excited  $(2^{3}S_{1})$  state, the atom is metastable as it cannot easily decay to its ground state due to symmetry restrictions. The large internal energy of 19.82 eV of this excited state is sufficient to ionize any atom it collides with, except for a helium atom in the ground state. This process, called Penning ionization, can be suppressed in a cloud of metastable helium atoms if the atoms are all polarized in the same maximum spin state. In this way clouds of ultracold metastable helium atoms can be prepared that decay on the timescale of tens of seconds, which provides enough time to do many types of experiments.

In our experiment we have the possibility to make ultracold metastable <sup>3</sup>He and <sup>4</sup>He atoms. These atoms only differ by a neutron in the nucleus, but this has a huge influence on their behavior when preparing an ultracold cloud with either atom. This has to do with their quantum statistical behavior. The <sup>3</sup>He atoms are fermions, and in an ensemble

of identical fermions no two fermions are allowed to occupy the same quantum state. As a result they are forced to occupy a large volume and energy distribution. If the cloud is cold enough the resulting distribution is very different from the behavior of a 'classical' gas and it is called a degenerate Fermi gas. The <sup>4</sup>He atoms are bosons and they behave exactly the opposite; they have no restrictions to having the same quantum state. In the right conditions (sufficient atom density at a low enough temperature) a trapped ultracold cloud of identical bosons will start to macroscopically occupy the lowest state of the system and form a so-called Bose-Einstein condensate. Both degenerate Fermi gases and Bose-Einstein condensates are exotic states of matter and allow investigation of fundamental atomic properties and quantum statistical behavior.

One of the experiments in this thesis, detailed in Chapter 3, focuses on the measurement of the transition frequency of the metastable <sup>4</sup>He atom to the so-called 2  ${}^{1}P_{1}$  state near the wavelength of 887 nm. Although extremely broad ( $\sim 290$  MHz), this transition is very weak and has never been directly observed before. Its measurement allows us to determine the energy of the 2  ${}^{1}P_{1}$  state with a relative accuracy of  $6.7 \times 10^{-10}$ . Comparing this value to calculations based on quantum electrodynamics, with a relative accuracy of  $4.8 \times 10^{-10}$ , shows a discrepancy of approximately three times the combined accuracy. This discrepancy is in agreement with two other measurements performed by a different group, and we can therefore assume that this discrepancy is not an experimental anomaly. There could be an issue with the theory, but as these calculations are very advanced there is no simple explanation to solve this issue. Apart from a determination of the energy of the  $2 {}^{1}P_{1}$  state, the measurement also provides a very accurate determination of the lifetime of the 2  ${}^{1}P_{1}$  state which is very short (~ 0.5 ns). This is a promising experimental feat as there have been many other measurements using completely different techniques, but this is the most accurate determination to date. As the theoretical prediction of lifetimes is orders of magnitude more accurate, this measurement does not provide a similar test of fundamental theory as it does for the energy of the  $2^1P_1$  state.

Chapter 4 focuses on the difference in quantum statistical behavior of a degenerate Fermi gas and a Bose-Einstein condensate. This difference is

observed by measuring the absorption line shape of a very weak transition from the metastable state to the (also metastable) 2  ${}^{1}S_{0}$  state near the wavelength of 1557 nm. Due to the completely different behavior of a degenerate Fermi gas and a Bose-Einstein condensate as explained before, the line shapes look completely different. The main difference is their spectral width, which differs by a factor five. This is a huge difference compared to a simple classical gas at an equivalent temperature, for which the spectral widths would be almost equal. We show that the line shapes (which also contain an asymmetry) can be fully understood by comparing them to an existing model and a model which we have extended ourselves. The measurements additionally provide a determination of an inter-atomic interaction parameter for the <sup>4</sup>He atoms which was never measured before, and is in agreement with new calculations based on theoretical models of the interaction potential between the atoms. This experiment is possible because of a new laser frequency stabilization scheme which employs state-of-the-art stabilization techniques involving a frequency comb and an ultrastable laser. As a result the spectral resolution of the laser system is good enough to observe these differences in line shapes, which was not possible before.

The transition from the metastable state to the 2  ${}^{1}S_{0}$  state is special because it can be calculated and measured to such precision, that the difference in nuclear size of the  ${}^{3}$ He and  ${}^{4}$ He atom (i.e. the size of the helion and the  $\alpha$ -particle) can be determined from a joint measurement in both atoms. This is a very interesting line of investigation, as similar measurements in muonic hydrogen and deuterium (neutral atoms in which the electron is replaced by a much heavier muon) have been done. The sizes of the proton and the deuteron determined in these experiments show a large discrepancy with the generally accepted values. As a next step similar experiments in  ${}^{3}\text{He}$  and  ${}^{4}\text{He}$  are necessary to see if there is a similar problem with slightly larger nuclei. A first measurement was done in our group in 2011, and Chapter 5 discusses several ways in which this measurement can be improved, so a better experimental accuracy can be achieved in the future. One improvement is gaining a better understanding of the line shapes of the atoms, as was discussed in Chapter 4. Chapter 5 discusses a major improvement which is called a 'magic wavelength trap' in which the measured transition frequency is no longer influenced by the trapping potential experienced by the atoms, and we calculate the possible candidate wavelengths for such a trap. Another improvement is a new method to accurately measure the local magnetic field which the atoms experience, as this also changes the measured transition frequency from the 'true' transition frequency. To conclude, this thesis discusses a couple of interesting topics that can be researched by measuring weak optical transitions in ultracold metastable helium. The excellent control and understanding of the behavior of the ultracold gas allows high-precision frequency metrology for testing fundamental physics such as quantum electrodynamics, quantum statistics and inter-atomic interactions, which are difficult or nearly impossible to measure using other methods.

### Samenvatting

Het helium atoom is een uitstekend testmodel voor fundamentele theorie. Het is een systeem dat in beginsel eenvoudig genoeg is om te begrijpen, maar de twee elektronen bieden meer complexiteit in de berekeningen vergeleken met het waterstof atoom, omdat het een drie-deeltjes systeem vormt in plaats van een twee-deeltjes systeem. Tevens zijn er ook extra elektron-elektron interacties. De meest nauwkeurige theorie tot op heden, kwantumelektrodynamica, kan de energieën van de verschillende lage elektronische toestanden van het helium atoom met 9 tot 10 cijfers nauwkeurigheid voorspellen (voor waterstof haalt het 11– 12 cijfers). Door deze voorspellingen te vergelijken met experimentele bepalingen, kan de theorie getest worden en in sommige gevallen informatie opleveren over atomaire eigenschappen, zoals de kerngroottes, die moeilijk op andere manieren te meten zijn.

Vanuit een experimenteel perspectief is het helium atoom zeer interessant. De stabiele elektronische configuratie van de grondtoestand maakt het lastig om het atoom met iets te laten reageren; het is een edelgas. Echter, in een aangeslagen toestand wordt het atoom zeer reactief. Als het atoom in de eerste aangeslagen  $(2 \ ^{3}S_{1})$  toestand zit, dan is het atoom metastabiel en kan het niet gemakkelijk vervallen naar de grondtoestand vanwege beperkingen in de symmetrie. De grote interne energie van 19.82 eV van deze aangeslagen toestand is voldoende om ieder atoom te ioniseren waar deze mee botst, behalve bij een ander helium atoom in de grondtoestand. Dit process, dat Penning ionisatie heet, kan onderdrukt worden in een wolk van metastabiele helium atomen als de atomen allemaal gepolariseerd zijn in dezelfde maximale spin toestand. Op deze manier kunnen wolken van ultrakoude metastabiele helium atomen geprepareerd worden die vervallen op een tijdschaal van tientallen seconden, wat voldoende tijd biedt om veel soorten experimenten te doen.

In ons experiment hebben we de mogelijkheid om ultrakoude metastabiele <sup>3</sup>He en <sup>4</sup>He atomen te maken. Deze atomen verschillen slechts met één neutron in de kern, maar dit heeft een enorme invloed op hun gedrag zodra een ultrakoude wolk met een van beide atomen geprepareerd wordt. Dit heeft te maken met hun kwantumstatistisch gedrag. De <sup>3</sup>He atomen zijn fermionen, en in een ensemble van identieke fermionen mogen geen twee fermionen een identieke kwantumtoestand hebben. Hierdoor worden ze geforceerd om een groot volume en grote energieverdeling in te nemen. Als de wolk koud genoeg is, zal de uiteindelijke verdeling heel anders zijn dan het gedrag van een 'klassiek' gas en heet het een ontaard Fermi gas. De <sup>4</sup>He atomen zijn bosonen en die gedragen zich precies het tegenovergestelde; ze hebben geen beperkingen om dezelfde kwantumtoestand te hebben. Onder de juiste omstandigheden (voldoende dichtheid bij een voldoende lage temperatuur) zal een ingevangen wolk ultrakoude identieke bosonen de laagste toestand van het systeem macroscopisch bezetten en een zogenaamd Bose-Einstein condensaat vormen. Zowel ontaarde Fermi gassen als Bose-Einstein condensaten zijn exotische toestanden van materie en kunnen gebruikt worden om fundamentele atomaire eigenschappen en kwantumstatistisch gedrag te onderzoeken.

Een van de experimenten in dit proefschrift, dat in Hoofdstuk 3 besproken wordt, concentreert zich op de meting van de overgangsfrequentie van een metastabiel <sup>4</sup>He atoom naar de zogenaamde 2  ${}^{1}P_{1}$  toestand bij een golflengte van 887 nm. Alhoewel deze overgang extreem breed is  $(\sim 290 \text{ MHz})$ , is deze erg zwak en nooit eerder rechtstreeks waargenomen. Met deze meting kunnen we de energie van de 2  ${}^{1}P_{1}$  toestand met een relatieve nauwkeurigheid van  $6.7 \times 10^{-10}$  bepalen. Als we dit vergelijken met berekeningen gebaseerd op kwantumelektrodynamica, met een relatieve nauwkeurigheid van  $4.8 \times 10^{-10}$ , zien we een verschil van ongeveer drie maal de gecombineerde onzekerheid. Dit verschil is in overeenstemming met metingen die door een andere groep gedaan zijn, en we kunnen daarom aannemen dat dit verschil geen experimentele afwijking is. Er kan een probleem zijn met de theorie, maar deze berekeningen zijn zeer geavanceerd en er is geen simpele verklaring om dit probleem op te lossen. Behalve de bepaling van de energie van de 2 $^{1}P_{1}$ toestand, heeft de meting ook een zeer nauwkeurige bepaling van de zeer korte levensduur van de 2  ${}^{1}P_{1}$  toestand (~ 0.5 ns) opgeleverd. Dit is een veelbelovend experimenteel resultaat omdat er veel andere metingen zijn, gebruikmakend van verschillende technieken, maar dit de meest nauwkeurige bepaling tot nu toe is. Omdat de theoretische voorspelling van de levensduur ordegroottes nauwkeuriger is dan het experimentele resultaat, kan deze meting niet gebruikt worden als test van de fundamentele theorie zoals dat voor de energie van de 2 $^{1}P_{1}$  toestand gedaan is.

Hoofdstuk 4 concentreert zich op het verschil in kwantumstatistisch gedrag tussen een ontaard Fermi gas en een Bose-Einstein condensaat. Dit verschil is geobserveerd door het absorptielijnprofiel te meten van een zeer zwakke overgang van de metastabiele toestand naar de (ook metastabiele) 2  ${}^{1}S_{0}$  toestand bij een golflengte van 1557 nm. Vanwege het eerder genoemde totaal verschillende gedrag van een ontaard Fermi gas en een Bose-Einstein condensaat, zijn de lijnprofielen zeer verschillend. Het voornaamste verschil is de spectrale breedte die een factor vijf verschilt. Dit is een enorm verschil vergeleken met een simpel klassiek gas bij een vergelijkbare temperatuur, waarvoor de spectrale breedtes bijna gelijk zouden zijn. We laten zien dat we de lijnprofielen (die ook een asymmetrie bevatten) volledig begrijpen door ze te vergelijken met een bestaand model en een model dat we zelf hebben uitgebreid. Bovendien kunnen de metingen gebruikt worden om een interatomaire interactie parameter te bepalen voor <sup>4</sup>He atomen die nog nooit eerder gemeten is. en in overeenstemming is met nieuwe berekeningen gebaseerd op theoretische modellen van de interactiepotentiaal tussen de atomen. Dit experiment is mogelijk vanwege een nieuwe laser frequentiestabilisatie methode met de allernieuwste stabilisatie technieken gebruikmakende van een frequentiekam en een ultrastabiele laser. Dit resulteert in een spectrale resolutie van het lasersysteem die goed genoeg is om deze verschillen in lijnprofielen te zien, wat nog niet eerder mogelijk was.

De overgang van de metastabiele toestand naar de 2  ${}^{1}S_{0}$  toestand is bijzonder omdat deze zo nauwkeurig berekend en gemeten kan worden, dat het verschil in de grootte tussen de atoomkernen van <sup>3</sup>He en <sup>4</sup>He (d.w.z. de grootte van het helion en het  $\alpha$ -deeltje) bepaald kan worden door een gezamenlijke meting in beide atomen. Dit is een zeer interessante lijn van onderzoek, omdat vergelijkbare metingen in muonisch waterstof en deuterium (neutrale atomen waarbij het elektron vervangen is door een veel zwaarder muon) gedaan zijn. De grootte van het proton en het deuteron die bepaald zijn in deze metingen, vertonen een groot verschil met de algemeen geaccepteerde waardes. Als volgende stap zijn metingen in <sup>3</sup>He en <sup>4</sup>He nodig om te zien of er zich een vergelijkbaar probleem voordoet bij grotere kernen. Een eerste meting is in 2011 gedaan, en Hoofdstuk 5 bespreekt verschillende manieren waarop deze meting verbeterd kan worden, zodat een betere experimentele nauwkeurigheid gehaald kan worden in de toekomst. Één verbetering is het beter begrijpen van de lijnprofielen van de atomen, zoals besproken in Hoofdstuk 4. Hoofdstuk 5 bespreekt een grote verbetering die ook wel een 'magische golflengte val' heet, waarin de gemeten overgangsfrequentie niet meer beïnvloed wordt door de valpotentiaal die de atomen voelen, en we rekenen de mogelijke kandidaat golflengtes uit voor zo'n val. Een andere verbetering is een nieuwe methode om nauwkeurig het lokale magnetisch veld te meten dat de atomen voelen, omdat dit ook de gemeten overgangsfrequentie verschuift.

Om samen te vatten: dit proefschrift behandelt een aantal interessante onderwerpen die onderzocht kunnen worden door zwakke optische overgangen te meten in ultrakoud metastabiel helium. De uitzonderlijke experimentele condities en het begrip van het gedrag van het ultrakoude gas staan ons toe om hoge-precisie frequentiemetrologie te doen. Hiermee kunnen we fundamentele natuurkunde zoals kwantumelektrodynamica en kwantumstatistiek toetsen, en ook inter-atomaire interacties bestuderen die lastig of bijna onmogelijk te doen zijn met andere methodes.

# Dankwoord

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