A magic wavelength optical dipole trap for high-precision spectroscopy of ultracold metastable helium



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ACADEMISCH PROEFSCHRIFT

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Ter nagedachtenis aan mijn vader

What might have been is an abstraction Remaining a perpetual possibility Only in a world of speculation.

T.S. Eliot

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Chapter 1

Background

Named after $H_{\lambda,oc}$, the greek titan god of the sun, the helium atom has played a starring role in the development of modern physics. The unraveling of its electronic structure pioneered by Heisenberg and perfected by Hylleraas marked the coming-of-age of quantum mechanics while its liquefaction by Kamerlingh Onnes kickstarted the field of low-temperature physics. The work in this thesis is built upon the impressive developments these fields have seen over the last century. In order to provide context to the present work, this chapter will give a brief overview of the current state of research in these fields. This chapter is structured as follows: section 1.1 introduces the field of ultracold atoms in general, and how it pertains to helium and precision spectroscopy in particular. Section 1.2 will give an overview of the theory of the helium atom, while section 1.3 gives a summary of experimental tests of this model. Section 1.4 introduces the proton radius puzzle and summarizes the current state of nuclear finite size measurements in helium which are the main motivations for the work in this thesis. Finally, section 1.5 outlines the work done as part of this thesis.

1.1 Ultracold atoms

In order to understand what is typically meant by the term "ultracold" in the context of atomic physics, consider a dilute gas comprised of particles that obey quantum mechanics. As the temperature of the gas decreases, the thermal de Broglie wavelength increases and the wave-like character of the particles becomes more apparent. For particles with a short range interaction potential, such as the van der Waals potential between atoms, the average wavelength of the particles can become much larger than the (effective) range of the interactions between them. In this limit the collision energy of two atoms is insufficient to support two-particle collisions with non-zero angular momentum and thus only collisions involving no angular momentum (s-wave) are allowed. In this limit, known as the s-wave limit, the scattering properties of atoms do not depend on the details of the molecular potential which can be effectively described as a zero-range hard-sphere potential characterized by its effective hard-sphere radius commonly referred to as the s-wave scattering length¹.

In the 1980s and 1990s, benefitting from improvements in laser technology, many techniques for cooling atoms were developed which allowed atomic gases to reach the s-wave regime. This culminated in the 1997 Nobel prize in physics which was awarded to Chu, Cohen-Tannoudji and Phillips [1–3] for their contributions to this field. Further innovations allowed cooling to even lower temperatures where the effects of quantum statistics dominate the behaviour of the gas. In this regime there is a profound distinction between particles with integer spin (bosons), and half-integer spin (fermions), which results in very different thermodynamic properties. For their observation of the associated phenomenon of Bose-Einstein condensation, a quantum phase transition originally predicted by Einstein [4], the 2001 Nobel prize in physics was awarded to Cornell, Wieman and Ketterle [5, 6].

Ultracold He^{*}

Today, research on cold atoms is a diverse and thriving field. Laser cooling techniques have been applied to many atomic species and even some molecules, pursuing a wide range of scientific goals. The species considered in this work, metastable helium (He^{*}), has some peculiarities that are not commonly encountered in other species [7]. Most of these stem from the fact that the atoms are in a metastable state which has a long natural lifetime of 8000 s and also (by atomic physics standards) an enormous internal energy of 19.82 eV. This internal energy is enough to ionize a collision partner in a process known as Penning ionization which must be suppressed by spin-polarizing the gas in order to reach appreciable densities (see section 2.1). On the other hand it allows for electronic detection methods which achieve a very high time resolution and signal to noise ratio compared to methods commonly used with other atomic species.

Due to its unique properties, He^{*} has been used in some remarkable experimental work. The first demonstrations of Bose-Einstein condensation of ⁴He [8, 9] were in fact already of considerable importance since, given the Penning-ionization problem, it was not obvious that this was even possible. Not much later the fermionic isotope ³He^{*} was also cooled to quantum degeneracy [10]. The electronic detection methods available for He^{*} have allowed a number of elegant demonstrations of effects originally studied in quantum

¹An important exception is the case of scattering between identical fermions for which scattering into partial waves with even angular momentum (which includes s-wave scattering) is forbidden.

optics. These include the Hanbury-Brown and Twiss effect for bosons and fermions [11, 12], the Hong-Ou-Mandel effect [13], and Wheeler's delayed choice experiment [14]. Besides at the VU in Amsterdam, there are groups actively engaged in research with ultracold He^{*} at the Institut d'Optique in Palaisau, Australian National University (ANU) in Canberra and at the university of Vienna.

Precision spectroscopy of cold atoms

Another important feature of He^{*} is that the electronic structure of helium is simple enough that very accurate ab-initio calculations are still feasible (see section 1.2). This is a unique feature for atoms that can be cooled to such low temperatures². Despite the absence of ab-initio calculations to test, precision spectroscopy of optical transitions in ultracold atoms is a very active field these days because these transitions can be used to realize extremely precise atomic clocks, beating the most precise Cs fountain clocks by orders of magnitude [19, 20]. Precision spectroscopy of helium stands to gain a lot from these developments. In this regard the group at the VU has played a pioneering role, measuring forbidden transitions in ultracold He^{*} [21, 22]. The work in this thesis is a continuation of that work, implementing an important technique used in optical lattice clocks known as the magic wavelength optical dipole trap (see section 1.5).

1.2 Theory of the helium atom

In contrast to the hydrogen atom, the non-relativistic Schrödinger equation for the helium atom is not analytically solveable. The reason for this is that the Hamiltonian is not factorizable with respect to the individual electrons and its two-electron eigenfunctions are not simply the product of single-electron wavefunctions. Nevertheless, even the earliest approximate methods that were developed during the formalization of modern quantum mechanics in the 1920s were quite succesfull in predicting properties of the helium atom where the old Bohr-Sommerfeld quantum theory had failed. For this reason the helium atom has been an important testing ground for theoretical methods and has played a pivotal role in the development of the description of multi-electron systems. Modern theoretical methods and computer power can now solve the non-relativistic system to essentially arbitrary precision.

 $^{^{2}}$ The simplest neutral atom, hydrogen, has been cooled to quantum degeneracy [15], but the associated experimental difficulty is such that it doesn't really benefit precision spectroscopy. Currently the most accurate spectroscopic measurements still rely on more basic atomic beam techniques [16–18].

1. Background

In order to develop insight into the helium atom it is useful to study the Schrödinger equation for a two-electron system in some detail. The equation must be solved for a wavefunction ψ in the coordinates \vec{r}_1 and \vec{r}_2 of the electron positions³ and is given by

$$\left[-\frac{\hbar}{2m_e}\left(\nabla_1^2 + \nabla_2^2\right) - \frac{e^2}{4\pi\epsilon_0}\left(\frac{Z}{|\vec{r_1}|} + \frac{Z}{|\vec{r_2}|} - \frac{1}{|\vec{r_1} - \vec{r_2}|}\right)\right]\psi(\vec{r_1}, \vec{r_2}) = E\psi(\vec{r_1}, \vec{r_2}),$$
(1.1)

where m_e is the electron mass and Z = 2 is the atomic number of helium. In a straightforward perturbative approach the Hamiltonian between the square brackets in equation 1.1 is divided into a separable part H_0 and an interaction part H_I . The general form is then

$$H_0 = \sum_{i=1}^{2} \left[-\frac{\hbar \nabla_i^2}{2m_e} - \frac{Ze^2}{4\pi\epsilon_0 |\vec{r_i}|} + V(r_i) \right], \qquad (1.2)$$

$$H_I = \frac{e^2}{4\pi\epsilon_0 |\vec{r_1} - \vec{r_2}|} - V(r_1) - V(r_2), \qquad (1.3)$$

where V(r) is an arbitrary central potential. The simplest choice is to omit this potential altogether (V(r) = 0) so that the eigenfunctions of H_0 are simple hydrogen-like wavefunctions. More accurate results are obtained with a central potential that phenomenologically incorporates screening of the nuclear charge by the other electron as was originally done by Heisenberg [23].

Regardless of the choice of central potential, the eigenfunctions of H_0 are products of single electron wavefunctions $u_{nlm}(r)$ which, like the eigenfunctions of the hydrogen atom, are spherically symmetric and characterized by quantum numbers n, l and m. Furthermore, the doubly excited states turn out to have energy higher than the helium ionization potential and are therefore unstable [24]. This means that the (anti-)symmetrized eigenfunctions of H_0 can be written as⁴

$$\psi_{\pm}(r_1, r_2) = \frac{1}{\sqrt{2}} \left(u_{100}(r_1) u_{nlm}(r_2) \pm u_{nlm}(r_1) u_{100}(r_2) \right).$$
(1.4)

The wavefunctions with positive sign are symmetric under the exchange of the two electrons and are collectively known as the *parahelium* states while the anti-symmetric wavefunctions are known as *orthohelium* states.

³Because of the spherical symmetry of the problem, the actual number of degrees of freedom is actually three instead of six. Variational basis sets often use Hylleraas coordinates $s = |\vec{r_1}| + |\vec{r_2}|$, $t = |\vec{r_1}| - |\vec{r_2}|$ and $u = |\vec{r_1} - \vec{r_2}|$.

⁴The only exception is the ground state wavefunction which is simply $\psi_+(r_1, r_2) = u_{100}(r_1)u_{100}(r_2)$.



Figure 1.1: Level scheme of the ⁴He atom. The parahelium states have total spin 0 (singlet) while the orthohelium states have total spin 1 (triplet) so that transitions between them are dipole forbidden. A number of optical transitions are indicated including the doubly forbidden $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition that is especially relevant in this thesis. The inset shows the fine-structure splitting of the $2 {}^{3}P$ state.

The full wavefunction including electron spin needs to be antisymmetric under the exchange of the electrons in order to satisfy the Pauli exclusion principle. The parahelium states must therefore be combined with an antisymmetric (singlet) spinor, while the orthohelium states have a symmetric (triplet) spinor. The effect of the perturbation Hamiltonian H_I is to lift the degeneracy between the ortho- and parahelium states, causing the latter to be somewhat higher in energy than the former. This can be seen in fig. 1.1 which shows the helium level scheme along with some of the important radiative transitions that can be observed.

An important consequence of the orthohelium-parahelium divide for spectroscopy is the fact that the dipole operator cannot change the spin state and therefore radiative transitions between orthohelium and parahelium are dipole forbidden. Furthermore, the dipole operator also requires a change in parity which does not occur for transitions between S-states. The practical consequence of this is that the first two excited 2 ${}^{1}S$ and 2 ${}^{3}S$ states cannot easily decay to the ground state and are metastable with natural lifetimes of 20 ms and 8000 s respectively. The next excited 2 ${}^{3}P$ state only decays into the 2 ${}^{3}S$ state, providing a closed transition.

Although it gives a physically intuitive picture, the perturbative approach is rather slow to converge because H_I is a rather large term. Modern calculations of the non-relativistic energy eigenvalues instead use a variational approach pioneered by Hylleraas [25–27]. With a properly chosen basis set and modern computer power the non-relativistic energies can nowadays be numerically evaluated to essentially arbitrary precision, as was perfected by Drake [28].

Higher order terms

Equation 1.1 is not yet a full description of the helium atom because a number of terms are still missing in the Hamiltonian. The first omission is that the nucleus is assumed to be stationary while in actuality the nucleus also moves in reponse to the movement of the electrons. This movement is very small however because of the small ratio of the electron mass to the mass of the nucleus $m_e/M(^{4}\text{He}) \approx 1.37 \times 10^{-4}$, or $m_e/M(^{3}\text{He}) \approx 1.82 \times 10^{-4}$ for ³He. The effect can therefore be accurately accounted for by expanding the Hamiltonian into powers of m_e/M . The terms in this expansion are called *recoil* correction terms.

The other omission is that equation 1.1 is, as mentioned, not correct in the relativistic limit. A physically intuitive justification of using the non-relativistic Hamiltonian is that the binding energy of the atom is small compared to the rest energy (i.e. mass) of the electron. This ratio is typically expressed in terms of the fine structure constant

$$\alpha = \sqrt{\frac{2hcR_{\infty}}{m_ec^2}} \approx \frac{1}{137}.$$
(1.5)

To correct for relativistic effects, the Hamiltonian can be expanded in powers of α . The leading order correction is the expectation value of the Breit-Pauli Hamiltonian [24] which in turn is derived from the Dirac equation. This term is the first to introduce a coupling term between the electron spins, the orbital angular momentum and the nuclear spin for ³He^{*}. As such it lifts the degeneracy between the different angular momentum states, producing a (hyper)fine structure splitting. This splitting is shown for the 2 ³P_{0,1,2} states in the inset of fig. 1.1. The theory of quantum electrodynamics (QED) predicts that the vacuum is not truly empty but rather that virtual particles can appear and disappear. The relativistic corrections beyond the first order must also consider interactions with these vacuum fluctuations. In the hydrogen atom these interactions lift the degeneracy between the 2 $S_{1/2}$ and 2 $P_{1/2}$ states which was measured by Lamb [29], providing the first experimental confirmation of QED theory. The QED corrections are therefore often called Lamb-shifts.

Taking into account all these interactions, the energy of any state of the helium atom can be written as a double expansion series in α and m_e/M according to

$$E_{tot} = hcR_{\infty} \sum_{i=0}^{\infty} \sum_{j=0}^{\infty} \alpha^{i} \left(\frac{m_{e}}{M}\right)^{j} \mathcal{E}_{(i,j)}.$$
 (1.6)

The $\mathcal{E}_{(0,0)}$ term corresponds to the non-relativistic energy and because $R_{\infty} \propto m_e \alpha^2$ this term is sometimes referred to as the $m_e \alpha^2$ order contribution⁵. The most advanced calculations of this type have been performed by Drake [30], and more recently by Pachucki and Yerokhin who have evaluated all terms in this series up to order $m_e \alpha^6$, $(m_e^2/M)\alpha^6$, $(m_e^3/M^2)\alpha^4$, and $(m_e^4/M^3)\alpha^2$ [31]. These calculations are typically accurate to a few MHz for the first excited states based on the estimated magnitude of the higher order terms.

Nuclear size shift

A final correction to the energies arises from the fact that the nucleus is not a point particle. Therefore, at distances shorter than the radius of the nucleus, the Coulomb potential felt by the electron is reduced. Because the radius of the nucleus $(R(^{4}\text{He}) \approx 1.681(4) \text{ fm} \text{ and } R(^{3}\text{He}) \approx 1.973(14) \text{ fm})$ is several orders of magnitude smaller than the Bohr radius $(a_0 \approx 52.9 \text{ pm})$ the electron density can be assumed constant over the nucleus. The nuclear finite size effect is then a contact interaction given by

$$\frac{Ze^2}{6\epsilon_0} \left\langle \sum_{i=1}^2 \delta(r_i) \right\rangle \left\langle R_N^2 \right\rangle = \frac{2\pi\hbar c Z\alpha}{3} \left\langle \sum_{i=1}^2 \delta(r_i) \right\rangle \left\langle R_N^2 \right\rangle. \tag{1.7}$$

The expectation value of the delta function operator characterizes the overlap of the electrons with the nucleus. This overlap is largest for S-states but very small for states with non-zero orbital angular momentum. For the S-states of hydrogen-like ions the overlap is given by

$$\langle \delta(r) \rangle = \frac{Z^3}{\pi n^3 a_0^3}.\tag{1.8}$$

⁵The first relativistic contribution is actually second order in α , corresponding to the $m_e \alpha^4$ term, from then on every order contributes.

From these equations it can be deduced that the 1S ground state of the hydrogen atom experiences a nuclear size shift of about 1.24 MHz ($r_p \approx 0.88$ fm). For the hydrogen-like He⁺-ion, with a larger nucleus and higher Z, this shift is about 71 MHz. The addition of another electron to make neutral helium adds another 30 MHz to this shift for the 1 ¹S ground state [31].

The nuclear size shift is dominantly a non-relativistic effect but it is smaller in magnitude than many of the high-order QED corrections. Additionally, high precision calculations must take a number of higher order corrections to equation 1.7 into account. One of these is the so-called two-photon exchange term [32] which is negligibly small for electronic helium but becomes the dominant theoretical uncertainty for muonic systems [33]. At present the theory of the helium atom is not yet accurate enough to resolve the nuclear structure directly but only via the isotope shift. Calculations of this shift can take advantage of the fact that operators that do not depend on the mass, radius or spin of the nucleus are common between different isotopes and can be neglected. The isotope shift can therefore be calculated to much higher precision allowing the extraction of the difference between the squared nuclear charge radii of different isotopes.

1.3 Comparison with experiment

A recent review by Pachucki compares precision spectroscopy experiments and QED calculations [31]. The 1 ${}^{1}S_{0}$ ground state ionization energy is challenging to accurately determine by experiment because of the deep UV light source that is required. The most accurate precision spectroscopy experiment starting from the ground state measured the transitions to the 4 ${}^{1}P_{1}$ and 5 ${}^{1}P_{1}$ states at 52.2 nm and 51.5 nm respectively [34]. The extracted ionization energy, accurate to 1×10^{-9} , is in agreement with theory but a factor 6 more accurate. Interestingly though, there is significant disagreement with the next most accurate determination using the 1 ${}^{1}S_{0} \rightarrow 2 {}^{1}S_{0}$ two-photon transition at 120 nm [35].

The long lifetime of the metastable 2 ${}^{1}S_{0}$ and 2 ${}^{3}S_{1}$ states (20 ms resp. 8000 s) provides a multitude of opportunities for precision spectroscopy in a more accessible wavelength range. Frequency metrology on many dipole allowed transitions has been performed, as shown in table 1.1. In addition two of the dipole-forbidden intercombination lines have been measured using the ultracold He^{*} setup at the VU. Except for 1 ${}^{1}S_{0} \rightarrow 2 {}^{1}S_{0}$ transition, the measured transition frequencies agree with QED calculations by Pachucki and Yerokhin. The ionization energies of the 3D levels which were not considered by Yerokhin and Pachucki but were calculated by the group of Drake to an accuracy of 20 kHz. Because of this high accuracy these energies are sometimes

Transition	Wavelength	Ref.
$1 {}^{1}S_{0} \rightarrow 4, 5 {}^{1}P_{1}$	52.2 nm and 51.5 nm	[34]
$1 \ {}^1S_0 \rightarrow 2 \ {}^1S_0$	$2{\times}120 \text{ nm}$	[35]
$2 {}^{3}S_{1} \rightarrow 2 {}^{3}P_{0,1,2}$	1083 nm	[36 - 38]
$2 {}^{1}S_{0} \rightarrow 2 {}^{1}P_{1}$	$2058~\mathrm{nm}$	[39]
$2 {}^{3}S_{1} \rightarrow 3 {}^{3}D_{1}$	$2{\times}762 \text{ nm}$	[40]
$2 {}^{1}S_{0} \rightarrow 3 {}^{1}D_{2}$	$2{\times}1009 \text{ nm}$	[41]
$2 {}^{3}P_{0} \rightarrow 3 {}^{3}D_{1}$	588 nm	[42]
$2 {}^{1}P_{1} \rightarrow 3 {}^{1}D_{2}$	668 nm	[43]
$2 {}^{3}S_{1} \rightarrow 2 {}^{1}P_{1}$	887 nm	[22]
$2 \ {}^3S_1 \rightarrow 2 \ {}^1S_0$	$1557~\mathrm{nm}$	[21], chapter 6

Table 1.1: Summary of high-precision spectroscopy measurements in the helium atom.

used as anchor points for determinations of the ionization energies of other levels. A number of recent experiments have found significant discrepancies involving these levels however, and Pachucki and Yerokhin have suggested that these calculations need further investigation.

Another important target for spectroscopy is the fine structure splitting of the 2 ${}^{3}P$ state. The different intervals have been measured using laser spectroscopy [44–46], as well as microwave spectroscopy [47, 48]. An important systematic effect in these measurements that for a long time was overlooked is quantum interference with far off-resonant transitions [49]. Correcting for this effect has resolved a number of discrepancies between different experiments. The fine structure intervals are especially sensitive to the value of the fine structure constant and have been used to determine this constant with an accuracy of 20 ppb [45], in agreement with the most accurate determinations [50, 51] and mainly limited by theory.

Other tests: Natural lifetime, polarizability and cold collisions

Besides energy levels theory predicts other properties of the helium atom that can also be tested experimentally. For example, the transition dipole moment between different states can be calculated, and the sum of these moments to lower states predicts the natural lifetime of a state. Two recent experiments have measured both the extremely long lifetime of the 2 ${}^{3}S_{1}$ state [52], and the extremely short 2 ${}^{1}P_{1}$ state natural lifetime [22], both in good agreement with theory.

A different property of the atom that can be calculated from theory is the linear response of an atom to an applied electric field, also known as the polarizability. One of the earliest tests of this kind was done using the tensor polarizability of the 2 ${}^{3}S_{1}$ state which produces an energy shift depending on

the relative orientation of the atom's spin and the electric fields. In 1968 the tensor polarizability of this state was predicted to appear as a result of relativistic corrections [53] and was measured shortly afterward [54] in good agreement with calculations. More recently, a tune-out wavelength, a wavelength where the total polarizability vanishes, for this state was predicted at 413 nm [55] which allowed a very sensitive test of polarizability calculations. This wavelength was measured to high accuracy [56] and advanced calculations including first-order relativistic and recoil corrections still found a significant discrepancy, indicating that the experiment is sensitive to QED correction terms.

In order to describe collisions between two or more helium atoms it is necessary to know their molecular potential, which can be calculated ab-initio. Laser cooling of He^{*} below the s-wave limit has allowed for very precise tests of these potentials. The least bound state of the ${}^{5}\Sigma_{g}^{+}$ molecular potential that describes collisions between spin polarized 2 ${}^{3}S_{1}$ atoms was measured to high accuracy using photo-association spectroscopy [57]. This measurement could then be used to finetune the ab-initio calculation of this molecular potential [58] and determine the s-wave scattering length to be 142.0(1) a_{0} . Other molecular potentials, which can become relevant in spin-mixtures, were calculated ab-initio by Müller *et al.* [59] with lower accuracy and have been used to make other predictions [60], including Feshbach resonances [61]. Experimental investigations of mixtures where these potentials are relevant have found no serious discrepancies [62, 63], but also no Feshbach resonances [64].

1.4 Measuring nuclear finite size effects

The nuclear finite size effect given by equation 1.7 is a small effect compared to many of the terms contributing to the Lamb shift. Nevertheless it has recently become a more prominent target for spectroscopy because of anomalous results in atomic hydrogen. This discrepancy has become known as the proton radius puzzle and has been a major motivation for the work on which this thesis is based. This section will introduce the puzzle, present the current state of nuclear-size related experiments in the helium atom, and describe the route to improvement that has been the driving idea behind the work in this thesis.

The proton radius puzzle

Due to the simplicity of the hydrogen atom its energy level structure can be calculated with extremely high precision [65]. These calculations involve several free parameters and in practice it is the experimental uncertainty in the determination of these parameters that limits the accuracy of the calculations. Of these parameters the Rydberg constant $R_{\infty} = \alpha^2 m_e c/2h$ is by far the most important but the proton radius r_p also contributes significantly to the uncertainty. The other parameters, the fine-structure constant α and the proton-electron mass ratio m_p/m_e , have a very minor effect because they can be determined to very high accuracy in independent experiments [50, 66].

Turning this problem around, one can assume that theory is correct which implies that R_{∞} and r_p can be determined via accurate spectroscopy of atomic hydrogen⁶. Such a measurement has important consequences for metrology because R_{∞} acts as a fulcrum for other physical constants. The most accurately measured transition in hydrogen (in fact, one of the most accurately measured transitions of any atom outside of atomic clock species) is the $1S_{1/2} \rightarrow 2S_{1/2}$ two-photon transition which has been measured with a relative accuracy of 4.2×10^{-15} [16, 67]. However, because the proton radius adds a second unknown to the problem, it is necessary to measure at least one other transition to fix both parameters.

Many other transitions in hydrogen have been measured [68–70] but none have achieved the same accuracy as the 1 $S_{1/2} \rightarrow 2 S_{1/2}$ transition. The accuracy on r_p obtained from elastic electron-proton scattering [71] is similar to the uncertainty from spectroscopy so it does not help much to constrain R_{∞} . An alternative to improving the accuracy of spectroscopy is to measure r_p to much higher accuracy so that it can be fixed. This was an important motivation for precision spectroscopy on the $2 S_{1/2} \rightarrow 2 P_{3/2}$ transition in muonic hydrogen (μH) where the electron is replaced by a muon. This elementary particle is identical to the electron apart from its far higher mass $(m_{\mu}/m_e \approx 207)$ which means that its wavefunction is much more confined and has a factor $(m_{\mu}/m_e)^3$ larger overlap with the nucleus (equation 1.8).

When researchers at the Paul Scherrer Institute in Villigen performed this ambitious experiment they found a surprise however. Using the Rydberg constant as input their measurements found an extremely accurate value for the proton radius that was significantly smaller than what was at the time the most accurately known value [72, 73]. This discrepancy has become known as the proton radius puzzle and, if confirmed, implies there is a difference between the muon and electron other than mass. This would be a direct violation of the principle of lepton universality which is one of the key assumptions in the standard model of physics. As of 2018, the puzzle remains unsolved with the latest CODATA recommended value for r_p (which explicitly excludes the muonic hy-

⁶In practice the Committee on Data for Science and Technology (CODATA) includes the transition frequency measurements as well as electron-proton scattering data as input data for a least squares fit for all physical constants together (including α and m_p/m_e) [66]. This is a somewhat opaque procedure because it is difficult to retrace how strongly different experiments contribute but it does yield the most accurate results provided that the input data is correct.



Figure 1.2: The proton radius puzzle. The muonic results are represented by the red line and uncertainty band, indicating the results from muonic hydrogen [72, 73], and the red poiont which indicates the result of muonic deuterium [74] corrected with the differential nuclear charge radius as measured in electronic hydrogen and deuterium [66]. The latest CODATA recommended value [66] (blue line and uncertainty band) explicitly excludes the muonic data and relies primarily on the combined electron-proton scattering data [75], and hydrogen spectroscopy (black points), H spect. is the CODATA regression excluding the e-p scatt. data. Note that the combined CODATA value is not simply the weighted average of these data because of other regression parameters. The green points are hydrogen spectroscopy measurements more recent than the latest CODATA recommended value [17, 18].

drogen result) disagreeing with the muonic hydrogen result at a significance of 5.6σ [66] as shown in figure 1.2.

The muonic hydrogen experiment and theory have been subjected to extensive scrutiny since the first publication. A follow-up experiment in muonic deuterium (μ D) also found a significantly smaller deuteron radius [74]. New, more accurate experiments in regular (non-muonic) hydrogen have been inconclusive, with one recent experiment finding agreement with μ H [17], while another confirms the CODATA value [18]. Furthermore, the analysis of world data for electron-proton scattering used by CODATA [75] is inconsistent with a "small" proton radius and although alternative analyses have found consistency with the μ H result [76–78], these are highly controversial [79].

The persistence of the proton radius puzzle has led to extensions of the research on muonic atoms to other species. Besides the already mentioned

measurements on μ D, experiments are also ongoing to measure the Lamb shift in the muonic helium ion (μ ^{3,4}He⁺), aiming to determine the radius of the alpha particle (⁴He nucleus) and the helion (³He nucleus) with sub-attometer accuracy [80]. The accuracy in this determination is expected to be limited not by the accuracy of the experiment but by the uncertainty in the calculation of the two-photon exchange term [33]. In order to find out if a "helium nucleus puzzle" exists this value will need to be compared with accurate experiments in regular (non-muonic) helium.

The size of the helium nucleus

Based on his analysis of the world data for elastic electron scattering, Sick derived the rms charge radii of the ⁴He and ³He nuclei (1.681(4) fm resp. 1.973(14) fm) [81]. The total nuclear finite size shift corresponding to these radii is smaller than the uncertainty in the most accurate QED calculations of transition frequencies but becomes the dominant source of uncertainty in calculations of the isotope shift. By measuring the isotope shift and using the accurately known radius of the ⁴He nucleus as an anchor point the charge radii of other isotopes can be determined.

A number of different experiments have measured isotope shifts to determine differential nuclear charge radius. One such measurement was performed at Argonne National Lab in Illinois using the exotic radioactive isotopes ⁶He and ⁸He (half lives 807 ms resp. 119 ms) on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{0,1,2}$ transitions [82, 83]. These experiments found that the nuclei have small charge radii (mostly determined by protons) compared to their total "matter" radii (protons and neutrons). This indicates that the neutrons are located relatively far from the nuclear center compared to the protons, a feature characteristic of so-called "halo" nuclei.

The (squared) nuclear charge radius difference between the stable isotopes ³He and ⁴He as determined by electron scattering is 1.066 ± 0.06 fm² [81] but can be measured to considerably higher accuracy in spectroscopic measurements of the isotope shift. The first experiments of this kind measured the isotope shift on the 2 ${}^{3}S_{1} \rightarrow 2 \, {}^{3}P_{0,1,2}$ transitions. The nuclear finite size effect on the isotope shift for this transition is 1212.2(1) kHz/fm² compared to the theoretical centroid frequency for a point-like nucleus which has been calculated to an accuracy of 0.9 kHz [31]. The accuracy realized in the most precise experiments on this transition is still experimentally limited however.

Shiner *et al.* measured the difference between the $2 {}^{3}S_{1} \rightarrow 2 {}^{3}P_{2}$ transition in ⁴He and the $2 {}^{3}S_{1}^{F=3/2} \rightarrow 2 {}^{3}P_{0}^{F=1/2}$ transition in ³He to an accuracy of 3 kHz [84]. Subtracting the hyperfine shift of the $2 {}^{3}S_{1}^{F=3/2}$ state [85], this difference can be compared to theory to yield the nuclear charge radius difference as $\delta r^{2}({}^{3}\text{He} - {}^{4}\text{He}) = 1.061(3) \text{ fm}^{2}$. Cancio Pastor *et al.* performed



Figure 1.3: Several isotope shift measurements determining the ³He to ⁴He nuclear charge radius difference. All points are calculated according to the most recent isotope shift calculations of Pachucki [31]. The blue points (Shin95, CP12, and Zheng17, resp. [37, 38, 84]) are measurements of the 2 ³S₁ \rightarrow 2 ³P_{0,1,2} centroid frequency, while the red points (vR11 and Reng18, resp. [21] and this work (chapter 6)) are measurements of the 2 ³S₁ \rightarrow 2 ¹S₀ transition frequency. The grey error bar (CREMA [33]) indicates the expected error bar for muonic helium (the actual value has not yet been published). For clarity, the much less accurate value determined in electron scattering (1.066 ± 0.06 fm² [81]) is not shown. Note that: (*) Zheng17 uses the ³He transition frequency from CP12, (**) Reng18 uses the ³He transition frequency from vR11, and (†) for CREMA only the errorbar is shown as the actual value has not yet been published.

absolute optical frequency measurements on many different lines to determine the centroid frequency of the $2^{3}S \rightarrow 2^{3}P$ transitions for both isotopes, yielding $\delta r^{2}({}^{3}\text{He} - {}^{4}\text{He}) = 1.069(3) \text{ fm}^{2}$ [36, 37]. However, a recent measurement of the $2^{3}S \rightarrow 2^{3}P$ centroid frequency in ${}^{4}\text{He}$ found a 20σ discrepancy with the value of Cancio Pastor *et al.*, which strongly affects the determined nuclear charge radius difference [38].

A significant problem for these measurements is the natural lifetime of the 2 ${}^{3}P_{0,1,2}$ states, limiting the spectroscopic linewidth to 1.6 MHz and therefore the accuracy that can be attained experimentally. Another problem is quantum interference [49], which can cause line shifts of the order of the currently realized experimental accuracy. An attractive alternative is to measure the isotope shift on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition instead. Although this transition is less sensitive to nuclear size effects (214.66(2) kHz/fm²), the theoretical uncertainty is smaller (0.19 kHz for a point-nucleus) [31]. In addition the transition is far less susceptible to quantum interference effects and the 20 ms natural lifetime of the 2 ${}^{1}S_{0}$ state gives it a very high quality factor of 2.4×10^{13} . The downside

is that the transition is doubly-dipole forbidden and therefore extremely weak with an Einstein A coefficient $A_{12} = 9.1 \times 10^{-8} \text{ s}^{-1}$ [86].

The 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition was observed for the first time in the metastable helium setup at the VU by van Rooij *et al.* [21]. They measured the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency in ⁴He to an accuracy of 1.8 kHz, and the 2 ${}^{3}S_{1}^{F=3/2} \rightarrow 2 {}^{1}S_{0}^{F=1/2}$ transition in ³He to an accuracy of 1.5 kHz. Taking the difference and, like for the result of Shiner *et al.*, correcting for the hyperfine splitting of the 2 ${}^{3}S_{1}$ state in ³He [85], allows a determination of the squared nuclear charge radius difference. The value found in this way is $\delta r^{2}({}^{3}\text{He}-{}^{4}\text{He}) = 1.028(11) \text{ fm}^{2}$ which disagrees by several standard deviations with the results on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{0,1,2}$ transitions.

The discrepancy between van Rooij *et al.* [21] and Cancio Pastor *et al.* [37] is 3.6σ (originally found at 4.0σ , the discrepancy has decreased somewhat as a result of theory re-evaluations). The measurements by Zheng *et al.* [38] cast serious doubt on this discrepancy however, and complementary measurements in ³He are needed to resolve the problem. Nevertheless, this leaves the discrepancy between van Rooij *et al.* and Shiner *et al.* [84] which, at 2.9σ , is still significant. Provided that the discrepancy in electronic helium can be resolved, a comparison with muonic helium may provide more insight into the proton radius puzzle. The experimental work on μ ^{3,4}He⁺ has been completed but as of the time of writing theoretical work necessary to extract the nuclear charge radii is still ongoing. Figure 1.3 shows an overview of spectroscopic determinations of the differential nuclear charge radius including the expected uncertainty of the μ ^{3,4}He⁺ determination [33].

1.5 Outline of this thesis

The main scientific goal of the work described in this thesis has been to perform an improved measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency in order to investigate the discrepancy in the ${}^{3}\text{He}{}^{4}\text{He}$ nuclear charge radius difference and hopefully to contibute to new insight into the proton radius puzzle by providing an accurate benchmark to compare to the μ ${}^{3,4}\text{He}{}^{+}$ measurements. In order to achieve this goal, a number of improvements to the setup used by van Rooij *et al.* [21] were implemented to improve the experimental precision and to reduce systematic effects. The most significant of these improvements have been an extensive upgrade of the frequency metrology infrastructure, and the implementation of a magic wavelength optical dipole trap.

The remainder of this thesis will describe this work in detail and is structured as follows: Chapter 2 will explain the experimental setup in detail, describing the He^{*} cooling beamline as well as the tools used to probe the atoms. Particular attention is paid to parts of the setup that are new compared to ear-

1. Background

lier work on the setup. Chapters 3 through 6 were originally written as research articles and have either been published in various journals or are currently under review. Finally, chapter 7 summarizes the findings and their impact, and gives an outlook at work that still needs to be completed.

Frequency metrology infrastructure

Since the measurements of van Rooij *et al.* [21], there have been two important additions to the shared frequency metrology infrastructure at the VU. The first is a Cs-clock which has a higher accuracy at all timescales compared to the previously used Rb-clock. The second is an ultrastable laser which can act as a local oscillator in the optical frequency domain. Finally a new spectroscopy laser with a smaller intrinsic linewidth was purchased to drive the $2 {}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition. The laser was phase-locked to the ultrastable laser in a transfer lock setup using the new hardware, allowing more accurate absolute frequency measurements and strongly reducing the laser linewidth. This setup is described in section 2.5, and also in more detail by Notermans [87].

The higher spectral resolution provided by a more narrow spectroscopy laser allowed for the observation of a number of interesting physical effects as described in chapter 3. The absorption lineshape was found to depend strongly on quantum statistics and these effects were enhanced by the ac-Stark shift. A published model for fermions provides good agreement with the observed lineshape for ³He, and for bosonic ⁴He a published lineshape model was extended to account for the ac-Stark shift effect. Finally, a Bragg-like scattering effect was observed to produce a splitting of the ⁴He absorption line.

A magic wavelength trap

The most important systematic uncertainty encountered by van Rooij *et al.* [21] was the trap induced ac-Stark shift. This uncertainty appeared because the long interaction time required to make the transition required the atoms to be held in an optical dipole trap during the excitation. Because the polarizability of the 2 ${}^{3}S_{1}$ level is different from that of the 2 ${}^{1}S_{0}$ level at the 1557 nm wavelength of the ODT used in that experiment, the trapping potential felt by the atoms is state dependent, causing a differential shift on the transition. Furthermore, because the atoms are distributed over a finite spatial extent inside the trap, the ac-Stark shift is position dependent, causing additional broadening of the absorption lineshape, as shown in chapter 3.

A solution to these problems while still satisfying the requirement that the atoms remain trapped during the excitation is to generate a trapping potential that is identical for both states. This is possible because the polarizabilities of both states independently vary as a function of trap laser wavelength. Therefore, at certain points the polarizabilities of both levels cross and the differential ac-Stark shift vanishes. These points are known as *magic wavelengths* and they find application in highly accurate atomic clocks [88, 89] which are now reaching accuracies far exceeding the best realizations of the SI-second [19, 20].

Chapter 4 describes calculations of the polarizabilities of the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ states and identifies a number of magic wavelengths on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition. The most promising candidate wavelength from the perspective of optical trapping was found at 319.815 nm. In order to generate a sufficiently deep trap at this wavelength a sufficiently intense laser source was required. Chapter 5 describes the laser system that was built for this purpose as well as the first demonstrations of optical trapping with this system.

A new measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition

Chapter 6 discusses an improved measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency in ⁴He that was performed using the innovations described in earlier chapters. The new measurement is an improvement over the result of van Rooij *et al.* [21] by an order of magnitude, and in addition accurate measurements were performed of the precise magic wavelength position and the scattering length between the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ states. The final chapter (chapter 7) gives a more detailed calculation and discussion of the extraction of the nuclear charge radius difference and gives a quick overview of new measurements in ³He which are currently underway at the VU. Finally, a number of improvements to the experiment are proposed that may allow for measurements of even higher accuracy.

Chapter 2

Experimental setup

The setup used in this thesis has been built up by several generations of PhD projects over the course of three decades. In order to provide context for the current work, this chapter will first give a brief historical overview of the history of the setup before discussing the technical details.

The metastable helium source, collimation, Zeeman slower and initial magneto-optical trap (MOT) were first built by Rooijakkers [90]. This setup was expanded by Tol [91] and Herschbach [92] who achieved a MOT of 10^9 atoms and performed the first evaporative cooling experiments. Koelemeij [93] investigated magneto-optical trapping at the 2 ${}^{3}S \rightarrow 3 {}^{3}P$ transition at 389 nm, in addition to the more common $2 {}^{3}S \rightarrow 2 {}^{3}P$ transition at 1083 nm, to improve the phase-space density in the MOT. Bose-Einstein condensation was first achieved by Tychkov [94] in an improved vacuum chamber and magnetic trap $(m_J = +1 \text{ atoms})$. Stas [95], McNamara[96], and Jeltes [97] built up a ³He^{*} recycling system and additional lasers, culminating in the realization of a degenerate Fermi gas of ³He^{*}, as well as of a doubly degenerate Bose-Fermi mixture of spin-polarized ⁴He^{*} and ³He^{*}. In a collaboration with the Institut d'Optique in Palaisau, this unique combination of having fermionic and bosonic species with a high internal energy allowed for an elegant demonstration of the Hanbury-Brown and Twiss effect for bosons and fermions in the same setup [12].

After this work, attention was shifted to frequency metrology. Roozendaal and van Rooij [98] implemented a more advanced computer program to control the experiment and realized an optical dipole trap (ODT) at 1557 nm. The ODT allowed trapping of the other magnetic substates of He^{*} and also proved to be a good platform for precision spectroscopy of the 2 ${}^{3}S \rightarrow 2 {}^{1}S$ transition. The transition was first observed and measured in this trap [21]. Significant improvements were made to the frequency metrology infrastructure by Notermans [87], which partly overlapped with the work presented in this thesis.

2.1 Laser cooling and magnetic trapping of He*

Optical setup

The optical setup was described in detail by Notermans [87]. Here I will give a brief overview. The light used for manipulating, cooling, and imaging the ⁴He^{*} atoms is derived from an ytterbium-doped fiber laser (NKT photonics). The laser is locked to the 2 ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{2}$ transition in a saturated absorption setup. Once the laser is locked, the light is amplified to about 700 mW by an ytterbium doped fiber amplifier (Nufern) and appropriately detuned and distributed by a series of AOM's as shown in figure 2.1. This setup provides separate laser beams with the following functions: collimation, magneto-optical trapping, optical pumping, Doppler cooling, slowing, and imaging.

For ³He^{*}, a separate laser, amplifier, and saturated absorption setup are used, locking instead to the 2 ${}^{3}S_{1}^{F=3/2} \rightarrow 2 \, {}^{3}P_{2}^{F=5/2}$ transition. The light from this setup is overlapped with the ⁴He^{*} light, providing identical detunings, power ratios, and alignment.



Figure 2.1: Optical setup to generate the laser beams required for collimating, cooling, trapping, and imaging He^{*}. Two separate lasers, saturated absorption setups, and amplifiers are used for both isotopes. These are overlapped in-fiber and passed through a network of accousto-optic modulators (AOMs) to provide the appropriate power and detuning. Typical powers used for ⁴He^{*} are indicated.

Source, collimation, and Zeeman slower

The He^{*} source is unchanged from the original design [90, 99]. It consists of a quartz tube with a tantalum needle electrode, a (non-conductive) boronnitride nozzle, and a skimmer 1 cm behind the nozzle. The tube is constantly being filled with helium while a DC-discharge is drawn between the needle and the skimmer. This discharge is typically run at a pressure of 2×10^{-5} mbar in the source chamber, a voltage of -2.7 kV, and a current of 7 mA. The boronnitride nozzle is cooled with liquid nitrogen to reduce the velocity of the beam exiting the skimmer to a most probable velocity of about 1100 m/s.

The beam coming from the source contains many atoms in states other than the desired 2 ${}^{3}S_{1}$, and also has a rather high divergence. Both of these problems are reduced by collimating and deflecting the beam. Resonant light is applied from four directions transverse to the beam with a wavefront curvature matching the trajectory of the atoms as they are being deflected. The light force applied to the atoms in this way is highly state selective and reduces the divergence of the beam considerably. To further improve the state-seclection, the beam is collimated under a slight angle so that they by-pass a knife edge that blocks the non-collimated parts of the beam.

When working with ${}^{3}\text{He}^{*}$, typically a mixture of ${}^{4}\text{He}^{*}$: ${}^{3}\text{He}^{*}$ at a ratio of 4:1 is supplied to the source and the output of the turbopumps of the collimation and source chamber are fed into a recycling system consisting of liquid nitrogen cooled molecular sieves. This system severely reduces the rate of consumption of the expensive and rare ${}^{3}\text{He}^{*}$ isotope.

After collimation and deflection, the atoms reach the "Zeeman slower" section of the beamline. Here the beam is illuminated by a circularly polarized laser beam from the opposing direction that optically pumps the atoms into the high-field seeking m = +1 state. The light force applied by this laser beam slows the atoms down but also pushes them out of resonance because of the reduced Doppler shift. To combat this, a spatially varying magnetic field is applied that induces a Zeeman shift exactly matching the reduction in doppler shift. At the end of the 2 meter long coil, the velocity of the atoms is reduced from 1100 to about 50 m/s, which is slow enough to capture them in a MOT.



Figure 2.2: Schematic side-view diagram of the He^{*} beamline. Metastables are produced in the source chamber from a DC-discharge. The atomic beam is then collimated by the interaction with a curved wavefront. The collimated beam is slowed from about 1100 m/s to about 50 m/s in the Zeeman slower. Finally, the slowed atoms can be captured in a MOT.

Penning ionization

The workhorses of cold-atom physics have, for a long time, been the wellestablished techniques of magneto-optical trapping (MOT) [100] and evaporative cooling [101]. The high (19.82 eV) internal energy of metastable helium presents a significant obstacle to the application of these techniques however. The reason is that the internal energy leads to losses via the exothermal reactions known as Penning ionization (PI), and associative ionization (AI)¹:

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

Together, these processes induce a two-body loss rate for unpolarized 2 ${}^{3}S_{1}$ atoms of ~ 10^{-10} cm³s⁻¹. In the presence of resonant laser light, this rate is enhanced even further to $10^{-8} - 10^{-9}$ cm³s⁻¹ by coupling to the 2 ${}^{3}P$ level, for which the Penning ionization-dominated two-body loss rate constant is ~ 10^{-7} cm³s⁻¹. This severely reduces the densities achievable in a MOT. Relatively large numbers of atoms can still be laser cooled however, by increasing the detuning from resonance and therefore the MOT volume [102].

By definition, a high atomic density is required to reach quantum degeneracy, and it would seem that the loss rates associated with Penning ionization render this completely unfeasible. It was realized early on however [103], that the Penning ionization rate (in the absence of resonant light) is reduced by four orders of magnitude in a spin-polarized gas of He^{*} atoms. The reason for this is that two spin-polarized ⁴He^{*} atoms (m = +1) have a total spin of 2, while the total spin of the reaction products cannot exceed 1 because of the spinless ground state of ⁴He. Eventually, spin conservation is broken by higher order terms (mainly due to the spin-dipole interaction) which do produce a finite coupling between Penning ionizing molecular potentials, but the loss rate associated ($\sim 2 \times 10^{-14} \text{ cm}^3 \text{s}^{-1}$) is no longer a hindrance for reaching high densities.

These considerations have far-reaching consequences for ultracold He^{*} experiments. The MOT stage, which requires cycling between different magnetic substates, should aim to limit the atomic density as much as possible. If, as is often the case, densities higher than those achieved in the MOT-stage are desired, the atoms should be prepared and remain in the spin-stretched m = +1 (or m = -1) state.

 $^{^1\}mathrm{In}$ practice, no distinction is made between these processes and both are referred to as "Penning ionization".

Magneto-optical and pure magnetic trap

After the Zeeman slower, the atoms enter the main chamber which is at ultrahigh vacuum (pressure 4×10^{-11} mbar or 1.0×10^{-10} mbar during operation). Here two coils in an anti-Helmholtz configuration generate a quadrupole magnetic field. The slow atoms entering this field are illuminated from six sides by circularly polarized laser beams, red-detuned from the $2 \ ^3S \rightarrow 2 \ ^3P$ transition. This configuration, which is known as a magneto-optical trap (MOT), causes the atoms to preferentially interact with the laser beam that pushes them back toward the quadrupole field center. Only in the center itself do the atoms interact with all beams equally in a configuration known as (red) "optical molasses", which cools the atoms.

In order to limit the density in the MOT and reduce losses via Penning ionization, the He^{*} MOT employs relatively large beams of 3 cm diameter (limited by the 2-inch optics used), and a relatively large detuning of -35 MHz. The quadrupole field applied by the anti-Helmholtz coils has an on-axis gradient of $\frac{\partial B}{\partial z} = 2 \frac{\partial B}{\partial \rho} = 22 \text{ G cm}^{-1}$. The MOT is typically loaded for about 4 seconds, yielding 4×10^8 atoms at a temperature of 0.56 mK. This configuration is chosen to optimize the number of atoms eventually cooled to quantum degeneracy. Table 2.1 gives an overview of some of the relevant experimental parameters at different stages of cooling.

When a sufficient number of atoms are loaded into the MOT, the cloud is transferred to a Ioffe-Pritchard cloverleaf-type magnetic trap which is described in detail by Tychkov [94]. Essentially, this trap creates a strong local minimum in the magnetic field which acts as a deep potential well for atoms with positive magnetic moment. The confinement in this trap can be made much higher than in the MOT and by employing the spin-stretched m = +1 state, the atoms are resistant to Penning ionization, allowing much higher densities. In order to optimize the transfer efficiency the magnetic trap is deliberately made more shallow using an additional power supply (the "small current" supply) and, as a side effect of this, has a relatively high field minimum of ~ 20 G. Furthermore the ⁴He^{*} (³He^{*}) atoms are optically pumped into the m = +1 (m=+3/2) state during the transfer using a 2 ms pulse of circularly polarized light.

After the transfer, the increased density and heating from optical pumping have raised the temperature compared to the MOT. To combat this, a laser beam is applied to perform 1D-Doppler cooling [104] for about three seconds. Because this process depends on reabsorption of spontaneously emitted light, the starting density needs to be sufficient. At this stage the trap typically contains about 1.8×10^8 atoms at a temperature of 0.13 mK. The density of the cloud is then further increased by reducing the current delivered by the second power supply which reduces the magnetic field minimum to ~1 G and compresses the magnetic trap.

2. Experimental setup

Table 2.1: Table of some of the relevant atomic properties and experimental parameters during different stages of cooling. All settings are optimized to produce the highest number of atoms at quantum degeneracy in the optical dipole trap (ODT). Transfer time between different stages is ≤ 0.5 seconds.

Laser cooling property	$^{4}\mathrm{He}^{*}$	$^{3}\mathrm{He}^{*}$				
Cooling transition	$2 {}^{3}S_{1} \rightarrow 2 {}^{3}P_{2}$	$2 {}^{3}S_{1}^{F=3/2} \rightarrow 2 {}^{3}P_{2}^{F=5/2}$				
Wavelength	1083.33064 nm	1083.46246 nm				
Natural linewdith $(\Gamma/2\pi)$	$1.62 \mathrm{~MHz}$					
Saturation intensity	$1.67 {\rm ~W} {\rm ~m}^{-2}$					
MOT						
Detuning	-35 MHz					
Gradient $\left(\frac{\partial B}{\partial z} = 2\frac{\partial B}{\partial \rho}\right)$	$22 \mathrm{~G~cm^{-1}}$					
Loading time	4 seconds	0.5 seconds				
Number of atoms (typical)	4×10^8	$6 \times 10^{7\dagger}$				
Temperature (typical)	$0.56 \mathrm{~mK}$	$0.6~{ m mK^{\dagger}}$				
1D Doppler cooling						
Doppler cooling time	3 seconds					
Number of atoms (typical)	$1.8 imes 10^8$	$6 \times 10^{7\dagger}$				
Temperature (typical)	$0.13 \mathrm{~mK}$	$0.3~{ m mK^{\dagger}}$				
Evaporative cooling and transfer to ODT						
Evaporation time	7.2 seconds	5.4 seconds				
Number of atoms (typical)	5×10^6	1×10^6				
Critical/Fermi Temperature	$T_c \approx 1.1 \ \mu \mathrm{K}$	$T_F \approx 1 \ \mu \mathrm{K}$				
Temperature (typical)	$0.2 \ \mu K$	$0.4 \ \mu { m K}$				

 \dagger Diagnostic reference measurements using 4 seconds of MOT loading leading to a greater amount of ${}^{3}\mathrm{He}^{*}$ captured. When sympathetically cooling with ${}^{4}\mathrm{He}^{*}$ the stated MOT loading time is used and less ${}^{3}\mathrm{He}^{*}$ atoms are captured.

At this high density collisions between atoms are frequent, allowing the atoms to quickly rethermalize after being brought out of thermal equilibrium. This allows forced evaporative cooling by applying a radiofrequency field that resonantly couples the m = +1 to the m = 0 state, causing them to be lost from the trap. The rf-frequency starts out very high (50 MHz) so that the only atoms resonant with the rf-field are at a high Zeeman shift and carry a high energy. After removing these high-energy atoms, less energy per atom is available and the energy distribution rethermalizes to a lower temperature. By ramping the rf-frequency down to ~ 3 MHz over the course of 7 seconds, the temperature is reduced to ~ 0.2 μ K and a Bose-Einstein condensate of $10^6 - 10^7$ atoms is produced.

For ³He^{*} in the spin-polarized hyperfine F = 3/2, $m_J = +3/2$ state, magnetic trapping and Penning ionization suppression work just as well as for ⁴He^{*}. However like all identical fermions spin-polarized ³He^{*} does not scatter via s-wave collisions. As a consequence, thermalization of an ultracold ³He^{*} is extremely slow compared to ⁴He^{*} and evaporative cooling of a pure ³He^{*} cloud is not possible. For this reason cooling of ³He^{*} is always performed in a mixture with ⁴He^{*}. During the evaporative cooling stage, the ³He^{*} atoms thermalize via collisions with ⁴He^{*} atoms, a process known as sympathetic cooling. This process is aided by the fairly large interspecies scattering length of 27 nm and the fact that due to its hyperfine structure the Zeeman splitting for ³He^{*} is smaller (by a factor 2/3) than for ⁴He^{*}, causing the losses from evaporative cooling to be born exclusively by the latter isotope. As a consequence much less ³He^{*} is needed in the MOT stage.

2.2 Detection

The main ultrahigh vacuum chamber has multiple means of detection in place. The most important of these are (1) absorption imaging, (2) the unbiased micro-channel plate (neutral MCP) detector, and (3) the biased ion-MCP. These different methods allow for the extraction of different but complementary information on the trapped gas. Absorption imaging is useful for probing of the atoms *in situ*, while the neutral MCP allows the measurement of a timeof-flight (TOF) profile with high time resolution, as well as a high signal to noise ratio. The ion-MCP measures ions instead of neutral atoms, which allows state-dependent and dynamic information. Below I give a description of the essential elements of each of these detection methods.

Absorption Imaging

Absorption imaging is a very versatile technique that is ubiquitous in coldatom experiments. The basic idea is to illuminate the atom cloud with a resonant laser beam and image the shadow cast by the atoms.

The imaging setup consists of a collimated laser beam that illuminates the atoms from the bottom². The beam is then imaged onto a camera via a one-to-one imaging setup. The InGaAs photodiode based camera (Xenics Xeva-1.7-320) has 320 by 256 pixels $30 \times 30 \ \mu\text{m}^2$ in size. The imaging beam has an intensity which is low compared to the saturation intensity ($I_{sat}/I \approx 10$) such that the Lambert-Beer law holds. The intensity profile falling onto the CCD

 $^{^{2}}$ When the neutral MCP is used, it blocks the imaging beam. When imaging it is moved out of the way via an in-vacuum translation stage.

is then related to the input intensity of the beam as

$$I_{out}(x,y) = I_{in}(x,y)e^{-\sigma_a n(x,y)},$$
(2.1)

where n(x, y) is the column density of the atoms, and σ_a is the (effective) photon absorption cross section. In order to be independent of the input intensity profile of the beam which may change over time, a second image, $I_{probe}(x, y)$ is taken shortly (typically 300 ms in the quantum degenerate regime) after the first image $I_{abs}(x, y)$ to provide a reference. The camera also picks up stray light and a pixel dependent dark current which is measured by taking a third image, $I_{bg}(x, y)$ with the imaging light blocked. The normalized transmission can now be calculated as

$$\frac{I_{out}(x,y)}{I_{in}(x,y)} = \frac{I_{abs}(x,y) - I_{bg}(x,y)}{I_{probe}(x,y) - I_{bg}(x,y)}.$$
(2.2)

Further improvement of the signal can be gained by using a "Fringe removal algorithm" [98]. This algorithm constructs a synthetic "optimal" $I_{probe}(x, y)$ as a linear combination of multiple previously obtained probe images. The optimal combination is found by fitting to the edges of the $I_{abs}(x, y)$ image, where no atoms are present. In practice this procedure improves the signal to noise ratio of the images, and also removes artifacts that result from distortions such as diffraction fringes in $I_{probe}(x, y)$ that are not reproduced in $I_{abs}(x, y)$. Figure 2.3 shows the different stages of image processing.

From the normalized transmission image the atomic column density is reconstructed as

$$n(x,y) = \frac{1}{\sigma_a} \ln\left(\frac{I_{out}(x,y)}{I_{in}(x,y)}\right).$$
(2.3)

Absorption imaging can thus in principle be used to determine the absolute number of atoms if σ_a is known exactly. In practice, performing a calibration of the MCP detector (see next subsection) turns out to be more reliable however, as the exact cross section depends on the details of temperature, light polarization, and internal state. See Tol [91] for a detailed analysis on this.

The main advantage of absorption imaging is that it allows *in-situ* imaging, providing an indispensible tool during the alignment of, among others, the optical dipole trap (ODT) beams. It also allows for a number of expansion measurements which are useful in characterizing the trap. One is the measurement of the trap frequencies by inducing an oscillation inside the trap, letting the atoms oscillate for a variable time, and observing in which direction they fly when the trap is suddenly switched off. The other is the measurement of relative populations in the three magnetic substates via a Stern-Gerlach measurement. See van Rooij [98] for detailed descriptions of these techniques.



Figure 2.3: Different image processing stages of an *in-situ* absorption image of a BEC trapped in the (320 nm) dipole trap. a) Raw absorption image. b) Corresponding probe image. c) Atomic column density recovered from these images according to equation 2.3. d) Atomic column density image including the fringe removal algorithm. The algorithm uses a total 139 probe images (including b)) taken on the same day.

Neutral MCP detection

The high internal energy of He^{*} atoms allows for detection methods not available to most other species. When a He^{*} atom hits a metal surface, its internal energy of 19.82 eV is more than enough to overcome the work function of the material, causing the release of an electron upon impact. This single electron can then be amplified in an electron multiplication process to produce a detectable current. This general process is the basis of many types of particle detector. He^{*} experiments typically use a micro-channel plate (MCP) detector because of its large active area and large number of channels.

After performing an experiment on a trapped cloud of atoms, the remaining atoms are released from the trap, allowing the cloud to expand and fall freely. These atoms can be detected if they hit an MCP detector (Hamamatsu F4655) located at a distance of 17 cm directly below the trap. This detector consists of an array of small electron multiplication channels with a high ($\sim 2 \text{ kV}$) voltage across them. The detector is shielded by a grounded grid so that no electric field emanates from the device, and the detector does not affect the path of particles falling onto it. Although in principle the detector is sensitive to any particle with sufficient energy, the device is meant to detect He^{*} atoms and is therefore referred to as the neutral MCP detector.

The high signal-to-noise ratio and time resolution of the MCP detector allow the measurement of the time-of-flight (TOF) profile of the ballistically expanding gas, from which many relevant experimental parameters can be deduced by fitting this signal with an appropriate fit function. The time-dependent signal on the MCP is given by

$$S_{MCP}(t) = N_{atoms} G\Phi(t), \qquad (2.4)$$

where N_{atoms} is the total number of atoms in the trap, and G is the net gain of the MCP (including quantum efficiency) which needs to be calibrated against a known number of atoms. The last time this was done [98], a gain of



Figure 2.4: Normalized flux $\Phi(t)$ according to the Maxwell-Boltzmann distribution at a temperature of 0.5 mK (blue line), 0.13 mK (orange line), and at 1 μ K (green line, decreased by a factor 1000 for clarity). Respectively, these temperatures roughly correspond to those achieved in the MOT, after Doppler cooling, and at the onset of quantum degeneracy where quantum statistics start to play a role.

 1.4×10^{-9} V s was found at an MCP voltage of 1.9 kV. The normalized flux of atoms $\Phi(t)$ represents the time dependent probability for a single atom to collide with the active surface of the MCP. Assuming the detector is positioned directly below the cloud it still depends on the height h = 17 cm of the trap above the MCP, the total active surface area which is circular with radius $r_0 = 7.25$ mm, and also, crucially, on the (thermodynamic) distribution of the atoms in the trap.

MCP flux models

The most basic thermal distribution of atoms in a trap is the Maxwell-Boltzmann distribution. For this distribution the normalized flux is given by [94]

$$\Phi_{MB}(t) = \frac{g}{\sqrt{2\pi}\sigma_{th}(t)} \frac{t^2 + t_0^2}{2t} \exp\left(-\frac{(h - gt^2/2)^2}{2\sigma_{th}(t)^2}\right) \left(1 - \exp\left(-\frac{r_0^2}{2\sigma_{th}(t)^2}\right)\right),\tag{2.5}$$

where g is the acceleration due to gravity, $\sigma_{th}(t) = t\sqrt{k_BT/m}$ is the size of the ballistically expanding cloud, and $t_0 = \sqrt{2h/g}$ is the free-fall time for a body initially at rest. Figure 2.4 shows the normalized flux for several temperatures commonly encountered in the He^{*} setup. The Maxwell-Boltzmann distribution is a good approximation of the flux when the phase space density is low. As the gas approaches quantum degeneracy (typically at $T \leq 1 \ \mu K$), the effects of quantum statistics start to become important, and the Bose-Einstein or Fermi-Dirac distribution must be used instead. Figure 2.5 shows the difference between these distributions. For bosons the normalized flux is given by [94]

$$\Phi_{Bose}(t) = g_3(z)^{-1} \frac{g}{\sqrt{2\pi}\sigma_{th}(t)} \frac{t^2 + t_0^2}{2t} \times \left[g_{5/2} \left(\zeta \exp\left(-\frac{(h - gt^2/2)^2}{2\sigma_{th}(t)^2}\right) \right) - g_{5/2} \left(\zeta \exp\left(-\frac{(h - gt^2/2)^2 + r_0^2}{2\sigma_{th}(t)^2}\right) \right) \right],$$
(2.6)

where $g_n(z) = \sum_{k=1}^{\infty} z^k / k^n$ are the so-called polylogarithm functions, and $\zeta = \exp(\mu/k_B T)$ is the fugacity of the gas, with μ the chemical potential of the (thermal) gas. For fermions, a Fermi-Dirac distribution is used instead which gives a flux expression which can be found by making the substitution $g_n(u) \rightarrow -g_n(-u)$ into equation 2.6. In the classical limit³ $\mu \rightarrow -\infty$ therefore $\zeta \rightarrow 0$. As the phase space density increases the chemical potential increases until it approaches zero (or a finite positive value for the degenerate Fermi gas).

For fermions, this expression remains valid in the limit $T \to 0$, where the chemical potential becomes equal to the Fermi energy ($\mu = E_F$) [105]. For bosons, the chemical potential tends to zero as $T \to 0$, and the point where it becomes lower than the lowest energy eigenstate of the system defines the critical temperature for Bose-Einstein condensation ($T_c \approx 0.94\hbar\bar{\omega}N^{1/3}$, where $\bar{\omega}$ is the average trap frequency, and N is the number of atoms [106]).

For interacting particles, the ideal Bose gas is not a good approximation below condensation, where the mean-field energy of the BEC must be considered. In our experiments, the BEC is well described by the Gross-Pitaevskii equation in the Thomas-Fermi limit [106], which gives a normalized flux of [94]

$$\Phi_{BEC}(t) = \frac{15}{16} g \frac{t^2 + t_0^2}{2t^2} \sqrt{\frac{m}{2\mu}} \max\left[0, 1 - \frac{m}{2\mu} \left(g \frac{t^2 - t_0^2}{2t}\right)^2\right]^2, \quad (2.7)$$

where for simplicity it is assumed that the entire BEC falls on the detector (in the He^{*} setup, this is ~ 95% if the BEC is directly above the detector). The TOF profile below T_c can be fit using a linear superposition of equation 2.6, and 2.7. The chemical potential (μ) in equation 2.7 is distinct from the chemical potential of the thermal gas (which is assumed to be zero below T_c) and is given by

$$\mu = \frac{1}{2} \left(15a\omega_x \omega_y \omega_z \hbar^2 m^{1/2} \right)^{2/5} N_{BEC}^{2/5}, \tag{2.8}$$

where ω_i are the trap frequencies in all three directions, and a = 7.512(5) nm or 142.0(1) a_0 is the s-wave scattering length [57]. This expression is very useful

³The thesis of Notermans [87] erroneously states the classical limit as $k_B T \gg \mu$ therefore $\zeta \rightarrow 1$.


Figure 2.5: Comparison of the momentum distribution for different kinds of particles. When quantum statistical effects are negligible the Maxwell-Boltzmann distribution (black solid line) is appropriate. When the phase space density increases, the energy distribution changes. For bosons, the Bose-Einstein distribution applies (blue dash-dotted line, $\mu = 0$), which gives a higher probability density at low momentum, an effect known as bunching. For fermions, the Fermi-Dirac distribution applies (red dashed line, $T/T_F = 0.25$), which shows anti-bunching.

because it relates the number of atoms in the BEC to the chemical potential which can now be measured directly by fitting equation 2.7 to measured TOF profiles. By inducing trap oscillations, the trap frequencies can be measured using absorption imaging, allowing a calibration of the MCP detector in terms of the absolute number of atoms [98].

Finally, when the temperature is significantly lower than the gravitational energy $(k_BT \ll mgh \approx k_B \times 0.8 \text{ mK})$, the movement of the atoms is dominated by gravity rather than ballistic expansion. This allows a simplification of expressions 2.6, and 2.7. For the Bose gas this gives

$$\Phi_{Bose}(t) = g_3(z)^{-1} \frac{g\sqrt{m}}{\sqrt{2\pi k_B T}} \times \left[g_{5/2} \left(\zeta \exp\left(-\frac{mg^2(t-t_0)^2}{2k_B T}\right) \right) - g_{5/2} \left(\zeta \exp\left(-\frac{mg^2((t-t_0)^2 + r_0^2/(2h)}{2k_B T}\right) \right) \right],$$
(2.9)

and for the BEC

$$\Phi_{BEC}(t) = \frac{15}{16} \sqrt{\frac{2\mu}{mg^2}} \max\left(0, 1 - 2\mu \frac{(t-t_0)^2}{mg^2}\right)^2.$$
 (2.10)

30



Figure 2.6: Time-of-flight profile of a BEC of 1×10^6 atoms with a thermal fraction of 5×10^5 atoms, fitted according to equations 2.9, and 2.10. The solid red line is the combined bimodal fit, while the black dashed line and the blue dotted line indicate the BEC and thermal Bose gas respectively.

Figure 2.6 shows a typical TOF-profile of a BEC with thermal fraction fitted according to equations 2.9 and 2.10.

Ion-MCP detection

Besides the neutral MCP, a second MCP detector is placed about 8 cm above the trap center (at a slight angle so as not to block any laser beams). This detector, known as the ion-MCP, is of the same type as the neutral MCP detector but doesn't have a grounded grid to shield the electric field emanating from it. This field affects the trajectory of nearby charged particles, attracting positively charged ions. The principle of detection is the same as for neutral particles except that the energy needed to release electrons is supplied by the potential on the front side of the MCP rather than by the internal energy of the He^{*} atoms. Because the flux of ions can be rather low compared to the flux on the neutral MCP it is sometimes advantageous to measure and count the individual current pulses on the ion-MCP rather than the integrated current.

The ion-MCP is a useful diagnostic tool because it allows the observation of dynamic processes during which the ionization rate changes as a function of time. Examples of such processes are MOT loading, and the onset of Bose-Einstein condensation which is associated with a sudden increase in density. The ionization rate also increases dramatically when the internal state of the He^{*} atoms is changed so that the cloud is no longer stable against Penning ionization. One way in which this happens is when the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition is made because for $2 {}^{1}S$ atoms Penning ionization is no longer suppressed. The ion-MCP thus allows the indirect detection of $2 {}^{1}S_{0}$ atoms.

2.3 Optical Dipole traps

Neutral atoms can also be trapped in laser beams which are far detuned from any resonances. The physical principle behind this is that the strong electric field induces a dipole moment in the atom, which allows the atoms to be trapped. Because of this induced dipole moment, such a trap is usually referred to as an optical dipole trap (ODT), although the terms far off-resonant trap (FORT) or quasi-electrostatice trap (QUEST) (when the laser beam is far reddetuned to the first electronic transition) are also sometimes used. Trapping atoms in a laser beam offers three important advantages over magnetic trapping. First, the trapping potential is independent on the magnetic substate, which allows the preparation of different spin-states, or even mixtures. Second, the high degree of control over a laser beam gives a lot of extra possibilities in shaping the potential landscape for the atoms at a microscopic scale. Finally, a laser beam based trap can be switched off much faster than a magnetic trapwhile minimally affecting the ballistic expansion of the gas.

Atomic polarizability

When an atom is placed in an electric field \vec{E} , the atom responds by forming a dipole moment \vec{p} . If the electric field changes in time, the dipole moment responds with some linear response function. In the Fourier domain

$$\vec{p}(\omega) = \alpha(\omega)\vec{E}(\omega), \qquad (2.11)$$

where $\alpha(\omega)$ is the linear response function of the dipole moment, better known as the polarizability. In an electric field oscillating at angular frequency ω , the atom then experiences an energy shift [107]

$$\Delta E = -\frac{1}{2} \langle \vec{p} \cdot \vec{E} \rangle = -\frac{1}{4} \operatorname{Re}(\alpha(\omega)) E_0^2, \qquad (2.12)$$

where E_0 is the peak amplitude of the electric field. This shift is known as the ac-Stark shift⁴. The atom also absorbs energy from the time-dependent electric field, which is re-emitted in the form of photons. The photon scattering rate is given by [108]

$$\Gamma_{sc} = \frac{P_{abs}}{\hbar\omega} = \frac{1}{2\hbar} \text{Im}(\alpha(\omega)) E_0^2.$$
(2.13)

 $^{^{4}}$ In the righthand side of equation 2.12 the root-mean-square value of the field was taken implicitly. For the dc-Stark shift it should be multiplied by 2.

The conservative part of the interaction, which is useful for trapping, is thus associated with the real part of the polarizability, while the dispersive part, which is detrimental to it, is associated with the imaginary part. The peak amplitude of the electric field of a monochromatic wave is related to the intensity of the wave as

$$E_0^2 = 2Z_0 I, (2.14)$$

where $Z_0 = 1/c\epsilon_0 \approx 376.7\Omega$ is the impedance of free space. A high intensity laser beam would thus provide an excellent trapping potential because of the high intensity gradient that can be generated. The only problem that remains is to choose a suitable frequency for the electromagnetic wave such that the real part of the atomic polarizability is large, while the imaginary part is small.

The Lorentz oscillator model

A simple classical model that reproduces much of the physics of the atomic polarizability is the Lorentz oscillator model [108]. This model treats the atom as consisting of a negatively charged "electron" that is elastically bound to a positively charged "nucleus", as if by a rubber band or spring. This system can oscillate with eigenfrequency ω_0 , and is damped by the emission of radiation. The corresponding on-resonance damping rate is

$$\Gamma = \frac{e^2 \omega_0^2}{6\pi\epsilon_0 m_e c^3},\tag{2.15}$$

where e, and m_e are the charge and mass of the electron respectively. When the system is driven by a linearly polarized electric field, the equation of motion is given by

$$\ddot{x} + (\omega/\omega_0)^2 \Gamma \dot{x} + \omega_0^2 x = -\frac{e}{m_e} E(t).$$
(2.16)

The dipole moment of this system p(t) = -ex(t) can be found by solving the equation of motion. By Fourier transformation

$$\frac{p(\omega)}{E(\omega)} = \alpha(\omega) = \frac{6\pi\epsilon_0 c^3}{\omega_0^2} \frac{\Gamma}{\omega_0^2 - \omega^2 - i(\omega^3/\omega_0^2)\Gamma},$$
(2.17)

where $e^2/m_e = 6\pi\epsilon_0 c^3\Gamma/\omega_0^2$ was substituted. Note that equation 2.17 only depends on the electron mass or charge to the extent that equation 2.15 does. The polarizability given by the Lorentz oscillator model provides a good approximation for real atoms that have a strong closed transiton. Γ should then be replaced by the spontaneous emission rate. The model fails when the response becomes non-linear, which is the case when saturation becomes non-negligible.

Plugging the polarizability of equation 2.17 into equations 2.12, and 2.13, the energy shift is given by

$$\Delta E = \frac{3\pi\epsilon_0 c^3}{4\omega_0^3} \frac{\Gamma}{\Delta} E_0^2, \qquad (2.18)$$

and the scattering rate by

$$\Gamma_{sc} = \frac{3\pi\epsilon_0 c^3}{4\hbar\omega_0^3} \left(\frac{\Gamma}{\Delta}\right)^2 E_0^2, \qquad (2.19)$$

where the rotating wave approximation was made, and $\Delta = \omega - \omega_0$ is the detuning from resonance. Two inferences can be made from these expressions which are very important to optical dipole traps. First, a red detuned ($\Delta < 0$) laser beam produces a negative energy shift, and thus provides a trapping potential, while blue detuned light ($\Delta > 0$) is anti-trapping. Secondly, the trapping depth (equation 2.18) decreases as Δ^{-1} , while the scattering rate (equation 2.19) decreases as Δ^{-2} . Because the latter induces heating and trap loss, it is advantageous to increase the detuning to improve the ratio of trap depth to loss rate.

Corrections to the polarizability

The Lorentz oscillator model is a good approximation of the polarizability when the laser is not too far detuned from a single strong line and the atom can be considered a two-level system. In general, other levels should be considered and the polarizability needs to be calculated from second-order perturbation theory [109]. The real part of the polarizability of some initial state n_i is then given by a sum over all final states n_f

$$\alpha_i(\omega) = -\frac{2}{\hbar} \sum_{f \neq i} \frac{\omega_{fi} |\langle n_i | \vec{d} | n_f \rangle|^2}{\omega_{fi}^2 - \omega^2}, \qquad (2.20)$$

where \vec{d} is the dipole operator, and ω_{fi} is the transition frequency between the final and initial state. This expression can be further rewritten in the LS coupling scheme as

$$\alpha_{n,J,m_J}(q,\omega) = 6\pi\epsilon_0 c^3 \sum_{n',J',m'_J} \begin{pmatrix} J & 1 & J' \\ -m_J & q & m'_J \end{pmatrix}^2 \frac{(2J'+1)A_{n'J'}}{\omega_{n'J'}^2(\omega_{n'J'}^2 - \omega^2)}, \quad (2.21)$$

where n, J, m_J are the principal, angular momentum, and magnetic projection quantum numbers respectively, and A_{nJ} is the Einstein A coefficient for the transition. The term between the large brackets is the Wigner 3j-symbol, and q is a quantum number for the polarization state of the light which is ± 1 , or 0 depending on whether the light has σ^{\pm} or π polarization respectively. In chapter 4 this formula is used for *ab-initio* calculations of the polarizability in order to find magic wavelengths.

In general the polarizability given by equation 2.21 depends on the exact magnetic substate, and the polarization of the light. It can be shown from symmetry considerations, that this dependence can always be parametrized as [89, 107]

$$\alpha_J(q,\omega) = \alpha_J^S(\omega) + (\hat{k} \cdot \hat{B})q \frac{m_J}{2J} \alpha_J^V(\omega) + (3|\hat{\epsilon} \cdot \hat{B}|^2 - 1) \frac{3m_J^2 - J(J+1)}{2J(2J-1)} \alpha_J^T(\omega),$$
(2.22)

where α_J^S , α_J^V , and α_J^T are known as the scalar, vector, and tensor parts of the polarizability respectively. The unit vectors \hat{k} , \hat{B} , and $\hat{\epsilon}$ are in the direction of, respectively, light propagation, the quantization axis (typically induced by an electric field), and the electric field polarization of the light. For the 2 3S_1 state of helium, the vector and tensor polarizability arise from perturbations to LS coupling [53] and are small enough compared to the scalar part that they can typically be neglected (see chapter 4).

Beam intensity profile

A diffraction limited laser beam, also known as a Gaussian beam, is described by a so-called TEM_{00} mode which, given a certain power P, has an intensity profile

$$I(r,z) = \frac{2P}{\pi w^2(z)} \exp\left(-\frac{2r^2}{w^2(z)}\right),$$
(2.23)

where the z coordinate is along the propagation direction and the r coordinate is orthogonal to it. The width of the beam $w(z) = w_0 \sqrt{1 + (z/z_R)^2}$ is fully characterized by the two related length-scales known as the beam waist w_0 , and the Rayleigh length

$$z_R = \frac{\pi w_0^2}{\lambda},\tag{2.24}$$

which characterizes the depth of focus. Far from the focus, the beam has a divergence angle $\Theta = \lambda/(\pi w_0)$ which decreases as wavelength decreases, making it easier to attain a small focus with short wavelength light. The perfect Gaussian beam is an idealization of real laser beams. In practice a laser beam may show a number of aberrations which tend to increase the minimum spot size achieved. This decreases the peak intensity, and therefore the trapping power of the beam, and for that reason minimization of these aberrations should receive consideration when selecting or building a trap laser. Nevertheless, the



Figure 2.7: Schematic view of the ODT geometry. The ODT beam is focussed in the vacuum chamber, refocussed, and passed into the vaccum chamber again, intersecting itself at an angle of 19° . The atoms are then trapped at the intersection.

Gaussian beam provides a good starting point for estimating the trapping potential for the atoms.

Crossed-beam ODT

In the He^{*} experiment, two laser systems are used to generate an ODT. The first is a commercial Erbium-doped fiber laser at 1557 nm (NP photonics Scorpio) which has a built-in amplifier with a maximum output power of about 2 W. The second is a home-built laser system at 320 nm which is described in detail in chapter 5, and produces up to 2 W at that wavelength. Both beams are focussed to have a similar waist size which is about 85 μ m for the 1557 nm laser, and about 60 μ m for the 320 nm laser, though with some astigmatism. The Rayleigh lengths of these beams are fairly long (~ 1.5 cm at 1557 nm, and ~ 3.5 cm at 320 nm) and provide very little confinement along the beam path. To remedy this, the beam is refocussed and passed into the chamber again, intersecting itself at an angle of $\theta = 19^{\circ}$. The polarization of the beam is rotated by 90° to prevent the generation of a standing wave. The geometry is shown in figure 2.7.

Assuming perfect Gaussian beams, what does the potential felt by the atoms look like? If the trap depth $(U_0 = \alpha I_{peak}/2\epsilon_0 c)$ is large compared to the temperature of the atoms, the potential is approximately harmonic with a slightly differing spring constant in three orthogonal directions [87]

$$U_{ODT}(x, y, z) \approx -U_0 \left(1 - \frac{m\omega_x^2 x^2}{2} - \frac{m\omega_y^2 y^2}{2} - \frac{m\omega_z^2 z^2}{2} \right), \qquad (2.25)$$

where the three trap frequencies are given by

$$\omega_x = \sqrt{\frac{4U_0}{mw_0^2}},\tag{2.26}$$

$$\omega_y = \omega_x \cos(\theta/2), \qquad (2.27)$$

$$\omega_z = \omega_x \sin(\theta/2). \tag{2.28}$$

The trap depth of the 1557 nm input beam has been estimated to be $U_0/k_B \approx 4.5 \ \mu\text{K}$ at an input power of 240 mW directly before the chamber [87]. For the 320 nm beam the peak intensity of a 1 W beam was found to be $1.0 \times 10^8 \text{W m}^{-2}$ (see chapter 5), and the polarizability to be $189.3 \times 4\pi\epsilon_0 a_0^3$ (see chapter 4). This provides almost the same trap depth at $U_0/k_B \approx 4.3 \ \mu\text{K}$. Based on this the expected trap frequencies are $(\omega_x, \omega_y, \omega_z) = 2\pi \times (359, 352, 59)$ Hz for the 1557 nm trap, and $(\omega_x, \omega_y, \omega_z) = 2\pi \times (500, 492, 82)$ Hz for the UV trap. The two trap frequencies ω_y , and ω_z can be measured using absorption imaging of trap oscillations. Typical trap frequencies found by this method are 50-80% of the expected frequencies. This mismatch can be attributed to misalignment of the beams (the alignment of the two waist positions is quite difficult to get right), and to beam aberrations that decrease the tightness of the focus.

Optical lattice

When the returning beam of the ODT has the same polarization as the incoming beam, the beams interfere and generate a standing wave pattern. The atoms feel this as a periodic potential and this configuration is known as an optical lattice [110]. This configuration has not been used in the experiments described in this thesis. However, a small unintended optical lattice was found to be responsible for the line-splitting effect found in chapter 3, and a deep optical lattice allows spectroscopy in the Lamb-Dicke regime which may be the next step in improving the measurements presented in chapter 6. This section therefore briefly reviews the basics of optical lattices, focussing on effects relevant to optical spectroscopy.

The 1D optical lattice potential can be written as

$$V_{latt}(x) = sE_{r,l}\cos^2(qx), \qquad (2.29)$$

where $s = U_0/E_r$ is the depth of the lattice in units of the lattice recoil⁵ $E_r = \hbar^2 q^2/2m$, $q = \pi/d$ is the lattice wavevector, and $d = \lambda/2\cos(\theta/2)$ is the lattice spacing. The recoil energy is the natural unit for measuring the lattice depth because it demarcates between the *weak potential* $(U_0/E_{r,l} \ll 1)$, and *tight-binding* $(U_0/E_{r,l} \gg 1)$ regimes of the optical lattice. Solving the Schrödinger equation (or Gross-Pitaevskii equation for a BEC) in the weak potential limit, one finds that a band-structure forms, with the two lowest bands given by (equation 19 of ref. [110])

$$\frac{E(q')}{E_{r,l}} = \left(\frac{q'}{q} - 1\right)^2 \pm \sqrt{4\left(\frac{q'}{q} - 1\right)^2 + \frac{s^2}{16}},\tag{2.30}$$

where q' is the quasi-momentum in the lattice. The main difference compared to free atoms is then the opening of an energy bandgap $\Delta = U_0/2$ at q' = q. This gap is unimportant as long as the momentum of the atoms does not come close to the lattice momentum, which is typically satisfied as long as the temperature is much lower than the recoil temperature $T_r = E_r/k_B$. However, when the recoil from a spectroscopy photon is close to the recoil of a lattice photon, which was the case for the experiments of chapter 3, even a small bandgap becomes noticeable and shows up as a splitting of the line.

The recoil energy⁶ of the 320 nm ODT ($E_r(320 \text{ nm}) \approx k_B \times 23.4 \mu \text{K}$) is much higher than for the 1557 nm ODT ($E_r(1557 \text{ nm}) \approx k_B \times 0.986 \mu \text{K}$), and therefore, in the weak potential limit, does not influence spectroscopy at 1557 nm. In the tight-binding limit, the lowest band can be described by [110]

$$\frac{E(q')}{E_r} = \sqrt{s} - 2J \cos\left(\frac{\pi q'}{q}\right), \qquad (2.31)$$

where the hopping parameter J, which is well-defined only in the tight-binding limit, is given by

$$J = \frac{4}{\sqrt{\pi}} s^{3/4} \exp(-2\sqrt{s}).$$
 (2.32)

In this limit, the bandgap is a multiple of the recoil energy, and excitation to the first excited band is forbidden by momentum conservation. At the same time, when the total width of the band becomes small compared to the recoil of a spectrocopy photon $(4JE_{r,l} < E_{r,s})$, excitation involving only the transfer of a single photon recoil is no longer possible. What is happening here is

⁵There can sometimes be some confusion over whether the recoil energy refers to the lattice or the spectroscopy photon recoil. An l or s, referring to the lattice or the spectroscopy, is added in the formulas to avoid ambiguity.

⁶These energies are for ⁴He, for ³He the recoil energies are higher at $E_r(320 \text{ nm}) \approx k_B \times 31.0 \ \mu\text{K}$, and $E_r(1557 \text{ nm}) \approx k_B \times 1.31 \ \mu\text{K}$.



Figure 2.8: Transition linewidth in the lowest energy band of an optical lattice at 320 nm as a function of $s = U_0/E_{r,l}$ compared to some energy scales relevant for spectroscopy on the $2^{3}S_1 \rightarrow 2^{1}S_0$ line in ³He^{*}. The blue dashed line is the bandwidth of the lowest energy band of the optical lattice, the grey dashed line is the recoil shift for ³He^{*}, the black dashed line is a typical Fermi temperature of 0.5 μK , the red dashed line is the natural lifetime of the 2 ${}^{1}S_0$ level, and the green dashed line is the off-resonant scattering rate at 320 nm. The purple solid line indicates the estimated linewidth for spectroscopy. The purple curve reaches a minimum of 13.9 Hz at s = 60.86.

that as the trap depth increases the atoms become more and more localized and confined to a single lattice site until they start to enter the Lamb-Dicke regime [111]. When the trap depth is increased further, at some point the bandwidth becomes smaller than the energy spread of the atoms, in particular for fermionic ³He^{*}. Eventually tunneling between different states is virtually impossible and the lattice becomes an array of unconnected harmonic potentials with trap frequency $\hbar\omega_0 = 2sE_{r,l}$, which is deep into the Lamb-Dicke regime when the spectrocopy recoil is comparable to or smaller than the lattice recoil.

These effects are relevant to spectroscopy of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition because they affect the recoil shift and the Doppler width of the absorption lineshape. Spectroscopic measurements should therefore aim to work either at $sE_{r,l} \ll E_{r,s}$ where these effects can be neglected, or at $sE_{r,l} \gg E_{r,s}$ to reap the benefits of the Lamb-Dicke regime. In the latter case, the proper lineshape should be calculated from Franck-Condon overlap integrals between the different Wannier wavefunctions that describe the atoms in this lattice. However, from a comparison of the energy distribution inside the trap and the expected bandwidth J, an estimate of the order of magnitude of the linewidth can be made. Figure 2.8 shows this comparison for the case of ³He^{*} in the 320 nm ODT. In the present configuration of the 320 nm ODT the maximum attainable trap depth is limited by the peak intensity of ~ 10^8 Wm⁻². At this intensity $s \approx 0.18$ and the experiment is far from the tight-binding regime. The only way of increasing the trap depth without compromising the magic wavelength condition would be to increase the light intensity, either by tighter focusing, or by using a build-up cavity. At some point however, lifetime broadening by off-resonant scattering, which also increases with intensity, becomes significant. Other loss mechanisms such as two-photon ionization may increase the lifetime broadening even further. If this is neglected there should be an optimum in the linewidth at around s = 60 where a linewidth less than 20 Hz is expected as can be seen in figure 2.8. This is a dramatic reduction compared to the linewidth of ³He^{*} which is limited by Fermi-energy of ~ $h \times 10$ kHz. For ⁴He^{*}, it may allow spectroscopy above the condensation threshold so that the density and thus the mean-field shift can be reduced.

2.4 Magnetic substate preparation

One of the advantages of the ODT compared to the magnetic trap is that the trap potential is (almost) independent of the magnetic substate of the atoms. This allows trapping of spin states other than the spin-stretched $m_J = +1$ state, or even spin mixtures. The rf-coil used for forced evaporative cooling, is also employed to drive transitions between different spin states. This section describes techniques by which these transitions are used to prepare different spin states.

Magnetic resonance rf-transitions

Because ⁴He has no nuclear spin, the Zeeman shift of the 2 ${}^{3}S_{1}$ state is given entirely by the electron spin. The Hamiltonian of this system⁷ with spin \vec{S} in a magnetic field \vec{B} is

$$\hat{H} = -\gamma \vec{B} \cdot \vec{S},\tag{2.33}$$

with the coupling given by the gyromagnetic ratio⁸ $\gamma = g_e \mu_B / \hbar \approx 2\pi \times 2.8025$ MHz G⁻¹, where μ_B is the Bohr magneton, and g_e is the electron *g*-factor. If the atoms are in a static magnetic field B_0 in the z-direction, and subjected to an oscillating field $B_1 \cos(\omega t)$ in the x-direction, equation 2.33

 $^{^7{\}rm This}$ Hamiltonian is readily generalized to systems with arbitrary angular momentum provided that the total angular momentum is a good quantum number.

 $^{^{8}}$ The 3 He gyromagnetic ratio is very close to 2/3 of this value, with only a minor correction due to the (much smaller) nuclear magnetic moment.

takes on the more explicit form

$$\hat{H} = \hbar\omega_0 \hat{S}_z + \frac{\hbar\Omega}{2} \left(e^{i\omega t} \hat{S}_- + e^{-i\omega t} \hat{S}_+ \right)$$
(2.34)

in the rotating wave approximation. Here $\omega_0 = \gamma B_0/\hbar$ is the Larmor frequency, $\Omega = \gamma B_1/(2\hbar)$ is the Rabi frequency, and \hat{S}_z and $\hat{S}_{\pm} = (\hat{S}_x \pm i\hat{S}_y)/2$ are spin projection operators. The time-dependent Schrödinger equation for this Hamiltonian, $\hat{H}|\psi(t)\rangle = i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle$, yields a set of coupled equations for the population amplitudes in the different spin states. These equations have equally spaced energy levels and idential diagonal matrix elements so we speak of a cascaded system.

⁴He^{*} is a spin-1 system with a three-dimensional Hilbert space. An arbitrary pure spin state of this system can be written as

$$|\psi\rangle = C_{+} |m_{J} = +1\rangle + C_{0} |m_{J} = 0\rangle + C_{-} |m_{J} = -1\rangle,$$
 (2.35)

where $|C_+|^2 + |C_0|^2 + |C_-|^2 = 1$ are the population amplitudes in the different spin projection eigenstates. Plugging the Hamiltonian of equation 2.34 into the time-dependent Schrödinger equation yields a system of three coupled differential equations for the population amplitudes

$$i\frac{dC_{+}}{dt} = \omega_0 C_{+}(t) + \frac{\Omega}{\sqrt{2}}\exp(-i\omega t)C_0(t)$$
 (2.36)

$$i\frac{dC_0}{dt} = \frac{\Omega}{\sqrt{2}}\exp(i\omega t)C_+(t) + \frac{\Omega}{\sqrt{2}}\exp(-i\omega t)C_-(t)$$
(2.37)

$$i\frac{dC_{-}}{dt} = \frac{\Omega}{\sqrt{2}}\exp(i\omega t)C_{0}(t) - \omega_{0}C_{-}(t).$$
(2.38)

The general solution to this system can be represented as a vector in the normalized orthogonal basis of solutions

$$|\psi_{0}\rangle = \frac{1}{2\Omega_{R}} \left(-\sqrt{2}\Omega e^{-i(\omega t + \phi)} |+\rangle + 2\Delta |0\rangle + \sqrt{2}\Omega e^{i(\omega t + \phi)} |-\rangle \right)$$
(2.39)

$$|\psi_{-}\rangle = \frac{e^{i\Omega_{R}(t-t_{0})}}{2\Omega_{R}} \left((\Omega_{R} - \Delta)e^{-i(\omega t + \phi)} |+\rangle - \sqrt{2}\Omega |0\rangle + (\Omega_{R} + \Delta)e^{i(\omega t + \phi)} |-\rangle \right)$$
(2.40)

$$|\psi_{+}\rangle = \frac{e^{-i\Omega_{R}(t-t_{0})}}{2\Omega_{R}} \left((\Omega_{R} + \Delta)e^{-i(\omega t+\phi)} |+\rangle + \sqrt{2}\Omega |0\rangle + (\Omega_{R} - \Delta)e^{i(\omega t+\phi)} |-\rangle \right).$$
(2.41)

where $\Delta/(2\pi)$ is the detuning from resonance, $\Omega_R = \sqrt{\Omega^2 + \Delta^2}$, and ϕ is the starting phase of the rf-field⁹. These solutions are eigenstates of the effective

⁹This phase is arbitrary for a single rf-pulse but becomes important when multiple pulses are used, such as in a Ramsey-type measurement.



Figure 2.9: Stern-Gerlach measurement of the magnetic field. The signal $n_-/(n_- + n_+)$ is determined from the absorption images above the graph where the different spin states are separated using a magnetic field gradient. The signal below is fitted using equations 2.45, and 2.43, where the Rabi frequency and the center frequency are the only free parameters. The Rabi frequency for this measurement is $2\pi \times 7.2$ kHz.

combined light-atom Hamiltonian¹⁰ in the dressed state picture with energies $E_{\pm} = \pm \hbar \Omega_R$ and $E_0 = 0$. The time evolution operator $U_{Rabi}(\Delta T, \phi)$ that evolves an arbitrary input state at time t_0 to the one at time t_1 is simply a projection onto this basis:

$$|\psi(t_1)\rangle = U_{Rabi}(t_1 - t_0, \phi_0) |\psi(t_0)\rangle = \sum_{i}^{+,0,-} |\psi_i(t_1 - t_0, \phi_0)\rangle \langle \psi_i(t_0, \phi_0) |\psi(t_0)\rangle.$$
(2.42)

Most of the time the system starts in the spin stretched state and the boundary condition $|\psi(t_0)\rangle = |m_J = +1\rangle$ is appropriate.

Rf-spectroscopy and Rabi-flopping

In the ODT, the atoms are still subjected to a magnetic field, either one that is applied, or the ambient field in the laboratory which is typically of the order of $\sim 0.5 G$. The associated Zeeman splitting can be measured by driving an rf-transition and observing the transfer to the different sublevels using a

¹⁰This Hamiltonian can be found by substituting $\omega_0 \rightarrow \Delta$ in equation 2.34

Stern-Gerlach type measurement (see section 2.2). The associated lineshape can be found using equation 2.42. Starting from the $|m_J = +1\rangle$ state, and applying an rf-pulse with excitation time T, the different populations are given by

$$|C_{+}(\Delta,T)|^{2} = \frac{\Delta^{2}}{\Omega_{R}^{2}} + \frac{\Omega^{2}}{\Omega_{R}^{2}}\cos^{4}\left(\frac{\Omega_{R}T}{2}\right) - \frac{\Omega^{2}\Delta^{2}}{\Omega_{R}^{4}}\sin^{4}\left(\frac{\Omega_{R}T}{2}\right)$$
(2.43)

$$|C_0(\Delta, T)|^2 = \frac{\Omega^2}{2\Omega_R^4} \left[\Omega_R^2 \sin^2(\Omega_R T) + 4\Delta^2 \sin^4\left(\frac{\Omega_R T}{2}\right) \right]$$
(2.44)

$$|C_{-}(\Delta,T)|^{2} = \frac{\Omega^{4}}{\Omega_{R}^{4}} \sin^{4}\left(\frac{\Omega_{R}T}{2}\right).$$
(2.45)

The Stern-Gerlach type experiment allows the measurement of relative spin state populations. Unfortunately, this measurement shows a strong negative bias in the relative $m_J = 0$ population because of the high rate of Penning ionization during the necessary expansion before imaging. Correcting for this bias is not straightforward because the effect is density dependent. Instead, the spectroscopy signal can be defined using only the relative population in the $|m_J = +1\rangle$, and the $|m_J = -1\rangle$ states as $S = n_-/(n_- + n_+)$. This signal can then be fitted according to equations 2.43, and 2.45, as shown in figure 2.9.

Equations 2.43-2.45 also describe the dynamics of Rabi flopping between the different magnetic substates, which can be used to transfer the atoms to different spin states. Not surprisingly, population transfer is most efficient when $\Delta = 0$, with a maximum of 50% transfer to the $|m_J = 0\rangle$ state for $\Omega_R T = \pi/2$, and 100% transfer to the $|m_J = -1\rangle$ state for $\Omega_R T = \pi$. Pulses with power and duration set to produce these population transfers are known as a $\pi/2$ - or π -pulse respectively. Figure 2.10 shows the measured population dynamics.

Landau-Zener sweep

A Rabi π -pulse is an efficient and fast way of transfering population from the $|m = +1\rangle$ to the $|m = -1\rangle$ state. A major downside however, is the fairly strong constraint on the detuning. Over the course of a single day the Larmor frequency has been observed to drift by several tens of kHz (~ 10 mG) which is significant compared to the maximum Rabi frequency (~ $2\pi \times 20$ kHz). A more robust method of population transfer is to use an adiabatic *Landau-Zener* sweep [112]. The basic idea behind this can be seen in figure 2.11 which shows the energies of as a function of detuning in the dressed state picture. Far from resonance the $m_J = +1$ state is identical to the $|\psi_+\rangle$ state. When the detuning is linearly ramped over the resonance (by sweeping the rf-frequency or the magnetic field) the spins remain in the $|\psi_+\rangle$ state, provided that the ramp is sufficiently slow. At the opposite side of the resonance the $|\psi_+\rangle$ projects almost



Figure 2.10: Observed population in different spin states, normalized to the total population at t=0, as a function of rf-pulse duration. The $m_J = +1, 0, -1$ populations are indicated by the blue squares, black circles, red diamonds respectively. The solid blue, dotted black, and dashed red lines are fits of equations 2.43-2.45 to the data. The measured $m_J = 0$ population is less than half the expected population because of Penning ionization losses. The Rabi frequency for this measurement is $2\pi \times 23$ kHz.

entirely into the $|m = -1\rangle$ state. The sweep therefore adiabatically transfers the atoms regardless of when it passes the resonance.

The problem that remains is to determine what ramping speed can be considered "sufficiently slow". An exact solution of the time-dependent Schrödinger equation during such a ramp is already quite complicated in the two-level system because the detuning Δ is now dependent on time. A useful approximation is given by the Landau-Zener formula [112] which gives the approximate probability of a non-adiabatic jump out of $|\psi_+\rangle$ (often referred to as the transfer probability)

$$P_{NA} = \exp\left(-\frac{2\pi\Omega^2}{\dot{\Delta}}\right). \tag{2.46}$$

The condition for adiabaticity is therefore $\dot{\Delta} \ll 2\pi\Omega^2$. A practical realization often used in the He^{*} setup [98] is to ramp the magnetic field from 1 to 2 G in 50 ms ($\dot{\Delta} = \gamma \Delta B/50$ ms $\approx (2\pi \times 3 \text{kHz})^2$), with a coupling Rabi frequency $\Omega \approx 2\pi \times 20$ kHz. With these numbers, equation 2.46 predicts an essentially perfect transfer to the $|m_J = -1\rangle$ state.



Figure 2.11: Schematic view of the energy levels $E_{\pm,0}$ in the dressed state picture during a Landau Zener sweep. At large detuning the eigenstates of the coupled system are the same as the uncoupled system. As the detuning continuously passes through zero the atoms remain in an eigenstate of the system which adiabatically transits into the opposite eigenstate of the uncoupled system.

2.5 Frequency Metrology

The experiments that form the core of chapters 3 and 6 rely heavily on the ability to accurately measure optical frequencies. To take care of the accuracy requirement, it is necessary that the measurement is traceable to the definition of the SI-second [113]:

The second is the duration of 9 192 631 770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of the caesium 133 atom.¹¹

The commercial Cs clock (Symmetricom CsIII Model 4310B) that is is used, is specified to be traceable to the SI-second to better than one part in 10^{12} . This is not accurate enough for the measurements described in chapter 6, so an additional calibration of the clock was done by comparing it to the time disseminated by GPS over long averaging time.

The Cs clock provides a frequency reference in the microwave domain ($\sim 10^{10}$ Hz), which is far lower than optical frequencies $(10^{14} - 10^{15}$ Hz). Since about twenty years, this gap can be bridged by making use of a special type of laser known as a frequency comb. This section describes the frequency comb and related infrastructure, as well as how these are employed to measure optical frequencies. The frequency comb and related infrastructure are maintained in close collaboration with the Ulrafast Laser Physics and Precision Metrology group of Kjeld Eikema.

 $^{^{11}}$ It was later made explicit that this statement refers to atoms not immersed in a thermal radiation bath i.e. at a temperature of 0 K [114].

Frequency comb

At the end of the 20th century, laser technology had progressed to the point that mode-locked oscillators could produce ultrashort (~ 100 ps) laser pulses in a phase-coherent pulse train. For their contributions to these developments, Hall and Hänsch were jointly awarded half of the 2005 Nobel prize¹² [115, 116]. To understand why these developments are considered so revolutionary, it is useful to consider the Dirac comb which is an idealization of the frequency comb. The Dirac comb consists of an infinitely long train of infinitesmally short "pulses" (i.e. delta functions) with a regular spacing T in time. Each pulse can have a cumulative phase offset ϕ and the total pulse train is normalized, so that the comb is given by

$$E(t) = \lim_{m \to \infty} \frac{1}{m} \sum_{m} e^{im\phi} \delta(t - mT).$$
(2.47)

The Fourier transform of this function is

$$E(\omega) = \int_{-\infty}^{+\infty} \lim_{m \to \infty} \frac{1}{m} \sum_{m} \delta(t - mT) e^{-i(\omega t - m\phi)} dt$$
(2.48)

$$= \lim_{m \to \infty} \frac{1}{m} \sum_{m} e^{-imT(\omega - \phi)} = \begin{cases} 1 \text{ for } \omega = \frac{1}{2\pi mT} + \phi \\ 0 \text{ otherwise} \end{cases}$$
(2.49)

The idealized comb thus consists of an infinite number of peaks in the frequency domain (called "modes"), with the frequency of the *n*-th mode given by

$$f_n = f_{ceo} + n \times f_{rep}, \tag{2.50}$$

where $f_{ceo} = 2\pi\phi$ is known as the carrier envelope offset, n is the mode number, and $f_{rep} = 1/T$ is the repetition rate of the comb. Figure 2.12 schematically shows the comb spectrum, both in the time and the frequency domain. Because f_{ceo} and f_{rep} are much lower in frequency than f_n , they can be referenced to an atomic clock so that f_n is known with the same (relative) accuracy. This allows the measurement of much higher frequencies with the same precision.

Of course in reality the pulse train is not infinitely long, and does not consist of pulses that are infinitely short. In practice a frequency comb only extends over a finite frequency range which is inversely proportional to the width of the pulses, while the finite coherence time of the laser causes a broadening of the modes. Nevertheless, the optical frequency comb has become an indispensible weapon in the physicist's arsenal.

 $^{^{12}\}mathrm{The}$ other half was awarded to Glauber.



Figure 2.12: Schematic view of the frequency comb in the frequency domain and the time domain. The frequency spectrum is given by equation 2.50 but only has appreciable power over a more narrow wavelength range limited by the inverse of the pulse time. The peaks have a finite width due to the loss of coherence over time. In the time domain the pulses are spaced a time T apart and carry a small carrier envelope offset phase ϕ_{ceo} .

Frequency combs have been generated at wavelength ranges extending from the mid-infrared [117, 118] to the extreme ultraviolet [34, 119], but the nearinfrared is still the most common. The frequency comb used for the experiments described in this thesis is a commercial erbium-doped fiber laser frequency comb (Menlo systems) which has a center wavelength of about 1500 nm and a bandwidth around 100 nm and is part of the shared frequency metrology infrastructure in the Atoms Molecules and Lasers group at the VU.

Ultrastable laser and transfer lock

The spectroscopy laser used in chapters 3 and 6 can be locked directly to the comb by generating a beat frequency and applying feedback to the comb. Unfortunately the linewidth of the comb modes is fairly large (~ 100 kHz, limited mainly by the stability of f_{ceo}) so this does not allow the full use of

the stability offered by the spectroscopy laser (NKT photonics E15) which has a specified short term linewidth of 0.1 kHz based on a delayed self-heterodyne measurement (delay of 120 μ s) [120].

This problem can be mitigated by using an ultrastable laser (ORS1500 by Menlo systems). In essence this is simply a continuous wave laser at 1542 nm locked to a reference cavity by the Pound-Drever-Hall technique. This reference cavity is made extremely stable however, using a number of techniques. The cavity is made out ultra-low expansion glass, is kept in high vacuum, temperature stabilized, and isolated from vibrations. With these stabilization techniques the specified laser linewidth is < 1 Hz, with a specified stability of 10^{-15} at 1 second. On longer timescales this stability becomes progressively worse as the laser slowly drifts.

The spectroscopy laser is phase-locked to the ultrastable laser by means of a transfer lock, using the frequency comb as a means of bridging the wavelength difference. The details and implementation of this technique are described in Notermans [87], here I give a brief overview. First, a beat note with the frequency comb is generated for both the spectroscopy laser and the ultrastale laser by overlapping each with light from the comb. These beat notes are converted into electronic signals using photodiodes. The electronic beatnote of the spectroscopy laser is mixed with an electronic signal from a direct digital synthesizer (DDS) in order to be able to tune the laser frequency later. This mixed beatnote is then mixed with the beatnote of the ultrastable laser and the frequency comb to generate the *virtual beatnote*. This virtual beatnote is compared to a local oscillator (the Cs clock) using a phase detector. This detector finally generates an error signal that is used to lock the spectroscopy laser. Figure 2.13 shows a schematic picture of this setup.

Because many of the effects that broaden the comb modes are common to the beat notes of the spectroscopy laser and the ultrastable laser, the contribution of the frequency comb to the final linewdith of the spectroscopy laser is strongly reduced. The carrier envelope offset frequency is entirely common between the two beat notes so any noise on this frequency does not contribute to the spectroscopy laser linewidth. The noise in the repetition frequency is suppressed by a factor $\Delta n/n$ which is the relative difference in modenumber between the two beatnotes. This reduces the comb contribution to the linewidth to about 1% of the linewidth of the comb modes (the absolute contribution to the linewidth of the spectroscopy laser is then about ~ 1 kHz). At present this is not a limiting factor to the experiments but if necessary it can be eliminated completely by employing more sophisticated mixing techniques [121]. Other contributions to the linewidth of the laser that were identified and estimated by Notermans [87] include the phase noise introduced by the ~ 80 m long fiber link to the He^{*} experiment (~ 2 kHz), and the electronic noise introduced by the analog lockbox used for the phase-locked loop ($\sim 1-2$ kHz). Based on



Figure 2.13: Schematic view of the transfer lock setup. The spectroscopy laser at 1557 nm, which is an erbium fiber laser (EFL), is mixed with the optical frequency comb (OFC), which is referenced to the Cs Clock, to generate a beatnote. An offset frequency generated by a direct digital synthesizer (DDS) is mixed into this beatnote to allow frequency steering of the spectroscopy laser. The ultrastable erbium fiber laser (UEFL) at 1542 nm is mixed with the same OFC to generate another beatnote which is mixed together with the spectroscopy laser beatnote, generating the virtual beatnote. The phase of the virtual beatnote is measured with respect to the Cs clock to generate an error signal that is used to lock the spectroscopy laser in a phase-locked loop (PLL).

these numbers, the total linewidth of the spectroscopy laser was estimated as 4-5 kHz at a timescale of 1 second.

Absolute frequency metrology

The frequencies of the spectroscopy laser, the ultrastable laser, and the virtual beat note can be written in reference to the frequency comb as

$$f_{sl} = f_{ceo} + n_{sl} f_{rep} + f_{b,sl} , \qquad (2.51)$$

$$f_{ul} = f_{ceo} + n_{ul} f_{rep} + f_{b.ul} , \qquad (2.52)$$

$$f_{b,virt} = f_{b,sl} - f_{b,ul} = f_{sl} - f_{ul} + (n_{sl} - n_{ul})f_{rep}$$
(2.53)

where n_i and $f_{b,i}$ are the modenumber and beat frequency for laser *i* with respect to the nearest comb mode (note that the f_{ceo} and f_b may have a negative sign). In order to measure an absolute laser frequency, it is first necessary to determine the modenumber of the nearest comb mode. This is done using a wavemeter with a resolution higher than the comb repetition rate. The absolute frequencies of both lasers can then be found by measuring (as well as determining the sign of) the comb carrier offset frequency f_{ceo} , the comb



Figure 2.14: Frequency measurement of the spectroscopy laser over the course of 12 hours. The graph on the left shows the reconstructed frequency (grey line), and its moving average at a timebase of 100 seconds (blue line). The red line is a linear fit to the data to compensate for the drift of the ultrastable laser. For this dataset the drift was 26(1) mHz/s. The graph on the right shows a histogram of the fit residuals.



Figure 2.15: The modified Allan deviation of the spectroscopy laser (blue squares), and of the same data with a linear drift removed (red circles). The red dotted line is a guide to the eye indicating the averaging rate of white frequency noise $(\tau^{-1/2})$.



Figure 2.16: Calibration of the Cs clock compared to the second disseminated by GPS. The graph on the left shows the time delay difference of the Cesium clock PPS compared to GPS PPS (grey line), and its moving average at a timebase of 10 000 seconds (blue line). The red solid line is a linear fit to the entire dataset and has a slope of $1.9(2) \times 10^{-14}$. The graph on the right is a histogram of the fit residuals, showing a slightly asymmetric distribution.



Figure 2.17: Allan deviation of the clock calibration data showing the modified Allan deviation of the Cs-GPS PPS comparison (blue circles), compared to the specified Cs clock stability (red squares). The dashed and dotted lines are guides to the eye, indicating averaging as τ^{-1} (blue dotted line), averaging as $\tau^{-1/2}$ (red dotted line), and the Cs clock stability floor of 5×10^{-14} (black dashed line).

repetition rate f_{rep} and, as the system is overdetermined, two of the three beat frequencies $f_{b,i}$.

These frequencies are measured using a four-port zero dead time counter (FXM50 by K+K Messtechnik) which is referenced to the Cs clock. Figure 2.14 shows the reconstructed frequency of the spectroscopy laser over a period of twelve hours. The frequency follows the ultrastable laser which drifts at a rate of 23 mHz/s on average. A linear fit to the data corrects for this drift. As the frequency measurements are averaged over a longer integration time, the uncertainty on the measurement reduces. The uncertainty of the measured frequency is typically given in terms of the (modified) Allan deviation [122]. Figure 2.15 shows the modified Allan deviation of the data shown in figure 2.14. The Allan deviation of the drift corrected signal diverges from the non-corrected signal at integration times longer than 1000 seconds (about 20 minutes), indicating that the drift of the ultrastable laser is significant at those timescales. The drift-corrected Allan deviation averages down to 2×10^{-13} after 12 hours, which corresponds to about 40 Hz on the absolute laser frequency. The driftcorrected Allan deviation matches the specified accuracy of the Cs clock at all investigated timescales, indicating that this is the limiting factor in the frequency determination.

All of these frequency measurements are done with respect to the frequency of the Cs clock. Although this frequency is specified to be stable to 5×10^{-14} , it is only traceable to the SI definition at the level of 10^{-12} [123]. In order to perform frequency measurements beyond this level it is necessary to calibrate the clock to a more accurate standard. A standard that can be used for this is the Global Positioning System (GPS) which disseminates a prediction of Universal Coordinated Time (UTC). Corrections to the GPS disseminated time are typically at the level of $< 10^{-15}$ when averaged over a one month period [124]. Figure 2.16 shows a comparison of the pulse-per-second signal (PPS) of the Cs clock, and GPS over a three-month period. This data is a lot noisier than the spectroscopy laser signal (figure 2.14) but the Allan deviation averages down faster which can be seen in figure 2.17, catching up with the stability of the clock at long integration time (> 10^6 seconds, or approximately 11 days). At integration time $> 10^5$ seconds (a little over 1 day) the Allan deviation averages down beyond 10^{-12} , allowing a calibration of the Cs clock. The time delay between pulses drifts at a rate of $\Delta T = (t_{Cs} - t_{GPS})/t_{GPS} = -1.9(2) \times 10^{-13}$, meaning that the Cs clock runs slightly faster than GPS. By correcting for this frequency offset the clock can be used to perform frequency measurements traceable to the SI-second up to its specified stability.

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Line shape of the $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition for bosons and fermions

Abstract

We observe a dramatic difference in optical line shapes of a ⁴He Bose-Einstein condensate and a ³He degenerate Fermi gas by measuring the 1557-nm 2 ³S - 2 ¹S 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 ⁴He a triplet-singlet swave 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.

3.1 Introduction

The bosonic or fermionic nature of a particle is a fundamental property, and trapped quantum degenerate gases display dramatic different behaviour 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

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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 ⁴He and a DFG of metastable ³He 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 [73, 74]. 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 [80, 125]. To determine the ³He-⁴He 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 [21]. The measured isotope shift, combined with QED calculations, allowed a determination of a squared nuclear charge radius difference of 1.028(11) fm² [32]. To compare this determination to measurements in muonic helium ions [80, 125] we aim to measure the $2^{3}S - 2^{1}S$ transition frequency with $\ll 1 \text{ kHz}$ 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.

3.2 Experimental setup

Our experimental setup is similar to earlier work [21] and to a more recent measurement of the 2 ${}^{3}S_{1} - 2 {}^{1}P_{1}$ transition at 887 nm [22]. We load a BEC of typically 10⁶ atoms in the metastable 2 ${}^{3}S_{1}$ ($m_{J} = +1$) state (lifetime ~ 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°, and the temperature of the thermal atoms in the ODT is typically $T \approx 0.2 \ \mu$ K. As the fermionic ³He atoms cannot thermalize once their temperature is below the p-wave barrier, they are loaded simultaneously with ⁴He and sympathetically cooled to degeneracy [10]. The quantum degenerate ³He-⁴He mixture is loaded into the ODT to rethermalize, after which the ⁴He atoms are blown away using resonant light. This procedure leaves a pure DFG of thermalized ³He 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 3.A). This is a factor 20 improvement compared to our previous experiment [21].

3.3 Comparing the line shape of a BEC to a degenerate Fermi gas

Figure 3.1 shows the optical σ^- 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⁻³. There are three times as many atoms in the BEC compared to the DFG due to the more complicated loading procedure of the DFG [10], 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 [126]) 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 ${}^{4}\text{He}$, 35 kHz for ${}^{3}\text{He}$). 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 [12]. For frequency metrology purposes it is clear that proper modeling is imperative in order to determine the true transition frequency.



Figure 3.1: 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 3.4). The zero frequency represents the transition frequency from the bottom of the trap.

The line shape for the DFG is calculated using the absorption line profile from Ref. [127] 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 $\mu/k_B = 0.55(15) \ \mu$ K of the DFG, the calculated line shape is shown in figure 3.1 (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 [15, 128]. Excellent agreement with the data was demonstrated, but the line shape function [129] cannot be used in our experiment for two reasons. First, in [129] 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 4), where s and t denote the singlet and triplet state $(2 \ ^1S$ atoms are repelled from the trap). Second, the excitation fraction in [129] 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 signal-to-noise ratio. Therefore we extend the Killian model [129], of which the full derivation is given in appendix 3.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 ${}^3S_1 - 2 {}^3S_1$ s-wave scattering length in the pure ${}^{5}\Sigma_{q}^{+}$ 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 date¹, a_{tt} is very accurately known: $a_{tt}^{\text{theory}} = 143.0(5) a_0$ [58, 130] and $a_{tt}^{exp} = 142.0(1) a_0$ [57], 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 3.A, is

$$S(\nu,\mu) = \frac{15\pi\hbar\Omega_R^2}{4} N \frac{h\nu}{\tilde{\mu}^2} \sqrt{1 - \frac{h\nu}{\tilde{\mu}}},$$
 (3.1)

where Ω_R is the Rabi frequency of the transition, N the total atom number of the BEC, ν 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 appendix 3.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

 $^{^{1}}A$ later measurement is described in chapter 6.

be written as

$$\frac{d\mu}{dt} = \frac{2}{5} \frac{\mu}{N} \tilde{S}(\nu, \mu) - \frac{2}{5} \Gamma \mu, \qquad (3.2)$$

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 3.A). We include the one-body lifetime Γ^{-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 equation 3.2 is numerically solved to fit to the line shape as shown in figure 3.1 (bottom). Here we use only the frequency offset and a_{ts} scattering length as free parameters, giving a reduced $\chi^2 = 0.94$.

3.4 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 $\lambda_{ODT} \approx 1557.3$ 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 photon in the lattice frame, $E_r = \hbar^2 q^{\prime 2}/2m \approx h \times$ 20.0 kHz, with $q' = 2\pi \cos(\theta/2)/\lambda_{\text{spec}}$, see figure 3.2(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 3.2(b). The resonance condition can only be satisfied below or above the bandgap, leading to a line splitting Δ of the transition, where $\Delta = V_0/2$ and V_0 is the optical lattice modulation amplitude as observed by the excited state [110, 131].

This excitation in a weak optical lattice is reminiscient of Bragg scattering of a BEC in an applied optical lattice [132, 133]. Contrary to Bragg scattering, where an applied moving optical lattice causes diffraction, the direct onephoton 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 3.3 shows that the splitting increases as V_0 is increased and in these measurements we estimate the optical lattice modulation amplitude



Figure 3.2: (a) In the crossed-beam optical dipole trap geometry we have a weak optical lattice with periodicity $d = \lambda_{\text{ODT}}/[2\cos(\theta/2)]$. Absorption of a spectroscopy photon results in a recoil momentum $q' = 2\pi \cos(\theta/2)/\lambda_{\text{spec}}$ in the lattice direction equal to the lattice recoil momentum π/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 $q' \approx \pi/d$ 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 Δ .

for the 2 ${}^{1}S$ state to be $V_0 \leq 6.5E_r^l$ 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 [134]. 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 3.3. For the DFG line shape measurements we have minimized V_0 by looking at the BEC spectra shown in figure 3.3. At this setting the DFG line shape is much broader than the effect of the lattice or, equivalently, the Fermi energy $E_F \gg V_0$.

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

We measure the time-dependent behaviour 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, and figure 3.4 shows the lines for 1 s and 3 s. The average reduced χ^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 1σ uncertainty based on simultaneous χ^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 appendix 3.B). Our result is in agreement with the estimated range of possible scattering lengths based on previous mean field shift measurements [21]. Furthermore the determination is in agreement with a surprisingly accurate theoretical value $a_{ts} = +42.5^{+0.5}_{-2.5}a_0$ [135], based on *ab initio* 1 ${}^{3}\Sigma_{g}^{+}$ and 2 ${}^{3}\Sigma_{g}^{+}$ molecular potentials [59] including large ionization widths which make the calculations insensitive to the actual coupling between the potentials. This is discussed in more detail in ref. [87].

3.6 Conclusion

To conclude, we have directly compared the fundamental difference between quantum degenerate fermions and bosons by measuring and calculating



Figure 3.3: 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 3.2(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 3.4: 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 3.1.

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 [127]. We extended the line shape of the Bose-Einstein condensate from the existing model [129] 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.

3.A Appendix: BEC lineshape model

Using the line shape model as calculated by Killian [129] as a starting point, the ac-Stark shift and the time-dependent behavior are introduced in two steps. Finally an analytic form of the line shape, convolved with a Lorentzian laser line shape, is given.

Adding an ac Stark shift

Although the hydrogen experiment was done in a magnetic trap [15, 128], 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 α 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, the potential for the excited state can be expressed simply as $V_2(\vec{r}) = (\alpha_2/\alpha_1)V_1(\vec{r})$, where α_1 and α_2 are the dynamic polarizabilities of the ground and excited states, respectively. In the main chapter the ground and excited states are labeled t and s respectively to distinguish between the singlet and triplet states.

The resonance condition of Killian already includes a spatially dependent mean field shift and the new spatially dependent term is simply added here. Using the general resonance condition

$$h(\nu - \nu_0) = V_2^{\text{eff}}(\vec{r}) - V_1^{\text{eff}}(\vec{r}), \qquad (3.3)$$

where $V_{1,2}^{\text{eff}}(\vec{r})$ are the effective potentials of the ground and excited state. Note that the definition of ν is slightly different from that used in equation 3.2 in section 3.3, which also contains the ac-Stark, Zeeman and recoil shift which are omitted here to simplify the notation. Filling in the mean field interaction and the ac Stark shift gives

$$h(\nu - \nu_0) = \left(\frac{\alpha_2}{\alpha_1} - 1\right) V_1(\vec{r}) + \Delta U n_1(\vec{r}).$$
(3.4)

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 $n_1(\vec{r})$. Inclusion of the ac-Stark effect thus 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. 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 μ 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.

There is a linear relationship between the ground state trapping potential and the density distribution of the BEC through the Thomas-Fermi relation



Figure 3.5: Schematic overview of the excitation in an energy-position graph. The unperturbed transition frequency ν_0 is ac-Stark shifted due to the different ac polarizabilities $\alpha_{1,2}$ of the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ states. In the Thomas-Fermi limit the Bose-Einstein condensate with chemical potential μ has a density profile $n_{\text{BEC}}(r,t)$ 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 ν_{\min} and ν_{\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.

 $\mu - U_0 = U_1 n_1(\vec{r}) + V_1(\vec{r})$, where $U_0 = V_1(r = 0)$ is the depth of the trap². Substituting this in equation 3.4 yields

$$h(\nu - \nu_0) = \left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) + \left(U_2 - \frac{\alpha_2}{\alpha_1}U_1\right)n_1(\vec{r}).$$
 (3.5)

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 equation 3.5 indeed reduces to the resonance condition as derived by Killian.

²The implicit definition $V_1(r \to \infty) = 0$ is convenient in these calculations but slightly unconventional as usually V(r = 0) = 0 is taken

The behaviour of the resonance condition can also be visualized in a schematic way as shown in figure 3.5. Outside of the optical dipole trap the resonance frequency is the unperturbed frequency ν_0 , but this frequency is ac-Stark-shifted in the optical dipole trap due to the differential polarizability of the states (as the polarizabilities $\alpha_2/\alpha_1 \approx -1.64$). Including the mean field interactions two extremes in the shifted transition frequencies are identified. The center frequency ν_{max} is given as the sum of the maximum ac-Stark shift and the maximum mean field shift $h\nu_{\text{max}} = [(\alpha_2/\alpha_1) - 1](\mu - U_0) + (U_2/U_1 - \alpha_2/\alpha_1)\mu$, using 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 > 0 the 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, the lineshape is given by

$$S(\nu) = \pi \hbar \Omega_R^2 \int 4\pi r^2 n_1(r) dr \times \delta \Big[h(\nu - \nu_0) + \Big(\frac{\alpha_2}{\alpha_1} - 1 \Big) (U_0 - \mu) - \Big(U_2 - \frac{\alpha_2}{\alpha_1} U_1 \Big) n_1(r) \Big], \quad (3.6)$$

where Ω_R is the Rabi frequency. This integral has the analytical solution

$$S(\nu) = \frac{15\pi\hbar\Omega_R^2 N}{4} \frac{h(\nu - \nu_0) - \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu)}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)^2 \mu^2} \times \sqrt{1 + \frac{h(\nu - \nu_0) - \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu)}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu}}, \quad (3.7)$$

with N the total number of atoms in the condensate. Equation 3.7 is also given in Section 3.3 as equation 3.2. This function is valid in the frequency domain

$$\left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) \le h(\nu - \nu_0) \le \left(\frac{\alpha_2}{\alpha_1} - 1\right)(\mu - U_0) + \left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu.$$
 (3.8)

This looks quite similar to equation 25 from Killian's work [129], 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 [129]) 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 the case considered here 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 μ and the atom number N of a BEC is the well-known nonlinear relationship

$$N = \frac{2^{\frac{5}{2}}}{15\sqrt{m}\hbar^2 a\bar{\omega}^3} \mu^{\frac{5}{2}},\tag{3.9}$$

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

$$\frac{dN}{dt} = \frac{dN}{d\mu}\frac{d\mu}{dt} = \frac{5}{2}\frac{2^{\frac{5}{2}}}{15\sqrt{m}\hbar^2 a\bar{\omega}^3}\mu^{\frac{3}{2}}\frac{d\mu}{dt} = \frac{5}{2}\frac{N}{\mu}\frac{d\mu}{dt}.$$
(3.10)

Realizing that the function $S(\nu)$ represents the one body atom number loss dN/dt, we can use this relationship to use equation 3.7 as a differential equation for the chemical potential:

$$\frac{d\mu}{dt} = \frac{2}{5} \frac{\mu}{N} S(\nu) = \frac{3\pi\hbar\Omega_R^2}{2} \frac{h(\nu - \nu_0) - \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu(t))}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)^2 \mu} \times \sqrt{1 + \frac{h(\nu - \nu_0) - \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu(t))}{\left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu(t)}}.$$
 (3.11)

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. To see if the condensate can redistribute fast enough during the excitation to assume equilibrium 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

$$\frac{1}{\mu}\frac{d\mu}{dt} = \frac{2}{5}\frac{1}{N}\frac{dN}{dt} \approx 0.13 \text{ s}^{-1},$$
(3.12)

assuming a (comparatively large) atom number of $N = 10^6$ atoms and a (comparativelt short) excitation time of 3 s based on the achieved depletion in figure 3.4 in section 3.5. The sound velocity is [136]

$$c = \sqrt{\frac{\mu}{m}} \approx 2 \text{ cm/s}, \qquad (3.13)$$

which, for a condensate with a length of ~ 200 μ m (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, it is reasonable to assume that the BEC remains 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 Γ , and the simple differential equation

$$\frac{dN}{dt} = -\Gamma N, \qquad (3.14)$$

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

$$\frac{d\mu}{dt} = -\frac{2}{5}\Gamma\mu. \tag{3.15}$$

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

Adding homogeneous broadening mechanisms

The spectral linewidth of the system (~ few kHz) is non-negligible compared to the observed width of the spectral feature (~ 15 kHz). In order to properly include a homogeneous broadening mechanism, the initial line shape is convolved with a broadening function $g(\nu)$ as

$$\tilde{S}(\nu) = \int_{\nu'} d\nu' g(\nu - \nu') S(\nu').$$
(3.16)

This does not change any of the first principle considerations, and a simple convolution of the analytical result for the lineshape with some arbitrary broadening mechanism (independent of the spatial distribution of the atoms) is in agreement with the formalism.

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

Defining the Lorentzian distribution as

$$g(\nu) = \frac{1}{2\pi} \frac{\gamma}{\nu^2 + (\gamma/2)^2},\tag{3.17}$$

with γ the full-width-half-max (FWHM), a closed form espression for the convolution with the line shape as defined in equation 3.7 can be obtained. Introducing the (rescaled) variables to simplify the final expression:

$$\tilde{\mu} \equiv \left(\frac{U_2}{U_1} - \frac{\alpha_2}{\alpha_1}\right)\mu,\tag{3.18}$$

$$h\tilde{\nu} \equiv h(\nu - \nu_0) + \left(\frac{\alpha_2}{\alpha_1} - 1\right)(U_0 - \mu),$$
 (3.19)

$$h\Delta \equiv \sqrt{(h\tilde{\nu} - \tilde{\mu})^2 + (h\gamma/2)^2},\tag{3.20}$$

$$h\delta_{\pm} \equiv h\Delta \pm (h\tilde{\nu} - \tilde{\mu}),$$
 (3.21)

$$h\xi_{\pm} \equiv \sqrt{(h\gamma)^2 \pm 8h\delta_{\pm}(h\tilde{\nu} - \tilde{\mu})}.$$
(3.22)

The line shape then becomes

$$\tilde{S}(\nu) = \frac{30}{32} \frac{\hbar \Omega_R^2 N}{\tilde{\mu}^{\frac{5}{2}}} \left[2h\gamma \sqrt{\tilde{\mu}} + \left(h\tilde{\nu}\sqrt{h\xi_-} + \frac{h\gamma}{2}\sqrt{h\xi_+} \right) \right] \\ \times \left(\tan^{-1} \left[\frac{-\sqrt{2\delta_+\tilde{\mu}/h}}{2\Delta - \sqrt{2\delta_-\tilde{\mu}/h}} \right] - \tan^{-1} \left[\frac{\sqrt{2\delta_+\tilde{\mu}/h}}{2\Delta + \sqrt{2\delta_-\tilde{\mu}/h}} \right] \right) \\ - \left(h\tilde{\nu}\sqrt{h\xi_+} - \frac{h\gamma}{2}\sqrt{h\xi_-} \right) \left(\log \left[\sqrt{\frac{h\delta_+\tilde{\mu}}{2(h\Delta)^2} + \left(1 - \sqrt{\frac{h\delta_-\tilde{\mu}}{2(h\Delta)^2}}\right)} \right] \right] \\ - \log \left[\sqrt{\frac{h\delta_+\tilde{\mu}}{2(h\Delta)^2} + \left(1 + \sqrt{\frac{h\delta_-\tilde{\mu}}{2(h\Delta)^2}}\right)} \right] \right) \right]. \quad (3.23)$$

This expression was used in equation 3.11 to calculate the time-dependent line shape used to fit the data as shown in figures 3.1, 3.3 and 3.4.

It is interesting to note that most of the parameters in the full analytical expression 3.23 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 Ω_{R} , as the overall prefactor of the lineshape contains the ratio $\Omega_{R}^{2}/\tilde{\mu}^{5/2}$ and using equation 3.18, there is a near-linear dependence between the scattering length a_{21} and Ω_{R} . The accuracy in determining a_{21} is therefore limited by the accuracy with which

the Rabi frequency can be estimated, which will be discussed in appendix 3.B. Note also that a number of assumptions were made in the derivation of this closed form expression (such as $\tilde{\mu} \ge 0$ which are true for the measurements in this work but not in general.

3.B Appendix: Estimating the Rabi frequency

Calculation of the Rabi frequency, which is an important parameter in our line shape calculations, requires trigonometry to calculate the projection of the spectroscopy beam on the quantization axis of the atoms. In the measurements used to determine the s-wave scattering length only the background magnetic field of the experiment was used. This section explains how this field is measured and how to calculate the Rabi frequency from this. A more detailed calculation can be found in ref. [87].

Consider a basic coordinate system where the z-axis is aligned with the axial direction (longitudinal axis) of the ODT and therefore of the BEC. The experiment is equipped with a set of coils in Helmholtz configuration ("fine tune coils") with their longitudinal axis parallel to this z-axis to apply a well-defined magnetic field if necessary. As the quantization axis of the atoms is given by the magnetic field, the magnetic field in this coordinate system can be calculated and used to determine the projection of the spectroscopy beam polarization to calculate the Rabi frequency.

Using rf spectroscopy to measure the magnitude of the magnetic field, as described in section 2.4, the fine tune coils are used to scan the magnetic field. The fine tune coils only produce a magnetic field component along the z-axis, and therefore these measurements can be used to determine the azimuthal angle $\theta = 52.1(4)^{\circ}$ of the background magnetic field with respect to the zaxis. With this angle we can calculate the projection P of the polarization of the spectroscopy light on the quantization axis. To be more precise, we are interested in the orthogonal part of the polarization $P' = \sqrt{1 - P^2}$ as this contributes to the σ^- -transition that is required to induce the 2 ${}^{3}S_1(m_J =$ $+1) \rightarrow 2 {}^{1}S_0(m_J = 0)$ transition. As the polar angle ϕ cannot be determined in the current setup, the full range is taken as a conservative estimate to be $P'^2 = 0.7 \pm 0.3$.

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

$$\Omega_R^2 = \frac{6\pi c^2}{\hbar\omega_0^3} A_{21} |\langle J_1 M_1 | q | J_2 M_2 \rangle|^2 I_0.$$
(3.24)

Here ω_0 is the transition frequency, $A_{21} = 9.1 \times 10^{-8} \text{ s}^{-1}$ the Einstein coefficient as calculated by Pachucki *et al.* [86], $I_0 = 2P_0/\pi w_0^2$ 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. σ^-, π, σ^+ , respectively), $C_{C-G}^2 = 1/3$ the corresponding Clebsch-Gordan coefficient of the transitions. Finally, an additional factor 1/2 needs to be added to account for the fact that the orthogonal component of the linear polarization decomposes equally into right- and lefthanded circularly polarized light.

Plugging all these numbers into equation 3.24 yields

$$\Omega_R = 2\pi \times (37 \pm 8) \text{ Hz}, \qquad (3.25)$$

where the uncertainty is fully dominated by the determination of P'^2 , and limits the systematic uncertainty in the determination of the s-wave scattering length. The circular component of the polarization due to the birefringent vacuum windows was judged to have an effect small compared to this uncertainty (see ref. [87]). Please note that a small error in the calculation was corrected giving a slightly higher estimate of the Rabi frequency compared to the value $\Omega_R = 2\pi \times (21 \pm 5)$ Hz found in ref. [87] and used in the published work [63]. This error has not been carried over into the data analysis and does not affect the conclusions of the main text in any way.

Chapter 4

Magic wavelengths for the $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition

Abstract

We have calculated ac polarizabilities of the 2 ${}^{3}S$ and 2 ${}^{1}S$ states of both 4 He and 3 He in the range 318 nm to 2.5 μ m and determined the magic wavelengths at which these polarizabilities are equal for either isotope. The calculations, only based on tables of level energies and Einstein A coefficients, do not require advanced theoretical techniques. Polarizability contributions of continuum states are calculated using a simple extrapolation beyond the ionization limit, yet the results agree to better than 1% with such advanced techniques. Several promising magic wavelengths are identified around 320 nm with sufficient accuracy to design an appropriate laser system. The extension of the calculations to ³He is complicated due to the additional hyperfine structure, but we show that the magic wavelength candidates around 320 nm are predominantly shifted by the isotope shift.

4.1 Introduction

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 [137], and spectroscopy in muonic hydrogen has allowed an even more accurate determination [72, 73]. Interestingly, the muonic hydrogen result currently differs by 7σ from the proton size determined by hydrogen spectroscopy and electron-proton collision experiments. So far

This chapter is based on: Magic wavelengths for the $2 \ ^3S \rightarrow 2 \ ^1S$ transition in helium, R.P.M.J.W. Notermans, R.J. Rengelink, K.A.H. van Leeuwen and W. Vassen, Physical Review A **90**, 052508 (2014).

there has not been a satisfying explanation for this discrepancy, which is aptly named the proton size puzzle [138]. Research in this field has expanded to measurements in muonic helium ions, a hydrogenic system which has a different nuclear charge radius [80]. As this work is done for both naturally occurring isotopes of helium (⁴He and ³He), the absolute charge radii of the α -particle and the helion may be determined at an aimed relative precision of 3×10^{-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 [139, 140]. Therefore isotope-shift 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 [21, 22, 34, 36, 37, 39, 43– 45, 48, 141] 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 [37] and the doubly-forbidden 2 ${}^{3}S \rightarrow 2 \, {}^{1}S$ transition at 1557 nm [21, 86] are measured at accuracies exceeding 10⁻¹¹, 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 4σ [37].

In order to determine the nuclear charge radius difference with a precision comparable to the muonic helium ion goal, we aim to measure the $2 {}^{3}S \rightarrow 2 {}^{1}S$ 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 [142]. 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.

In this paper we calculate the wavelength-dependent (ac) polarizabilities of both 2 ${}^{3}S$ and 2 ${}^{1}S$ states and identify wavelengths at which both are equal for either ⁴He or ³He. 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 [21] we employ a 1557 nm ODT at a power of a few 100 mW, providing a trap depth of a few μ K and a scattering lifetime of > 100 s (the actual lifetime in the trap is limited to 10's of seconds due to background collisions). A good overview on calculating trap depths and scattering rates in ODTs is given in [108], and the specific calculations for our ODT are discussed in section 2.3. 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.

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 [143, 144]. It is therefore sufficient for us to find magic wavelength candidates with an accuracy on the order of a few GHz, well within the tuning range. Although this is less accurate than polarizability calculations achievable with more advanced theoretical techniques [145, 146], the purpose of this paper is to show that it is possible to calculate magic wavelengths with sufficient accuracy to design an appropriate laser system without having to resort to the aforementioned specialized techniques.

The polarizabilities for the 2 ${}^{3}S$ and 2 ${}^{1}S$ states of ⁴He are presented over a wavelength range from 318 nm to 2.5 μ m. In this range all magic wavelengths including estimated ODT powers and 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 4.1 for an overview). Therefore these are used as a benchmark for our calculations by also calculating the polarizabilities in the dc limit ($\lambda \to \infty$), as discussed in section 4.4. Calculations of the ac polarizability of the 2 ${}^{3}S$ and 2 ${}^{1}S$ states [55, 147] states also allow us to compare the polarizability calculations at finite wavelengths.

Finally we present a simple extension to ³He which shows 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 ³He magic wavelength candidates and show that they are equal to the ⁴He results approximately shifted by the hyperfine and isotope shift.

4.2 Theory for ⁴He

The polarizability α of an atomic state with angular momentum J and magnetic projection M_J contains a scalar part α_0 , a vector part α_1 and a tensor part α_2 , all depending on J. In general, given a quantization axis \hat{B} , the polarizability in a laser field with propagation direction \hat{k} , polarization q(q = 0 for linear polarization, $q = \pm 1$ for σ^{\pm}), and electric field direction $\hat{\epsilon}$ is given by 1 [89, 107]

$$\alpha(\omega) = \alpha_0(\omega) + (\hat{k} \cdot \hat{B})q \frac{m_J}{2J} \alpha_1(\omega) + (3|\hat{\epsilon} \cdot \hat{B}|^2 - 1) \frac{3m_J^2 - J(J+1)}{2J(2J-1)} \alpha_2(\omega), \quad (4.1)$$

The non-scalar terms in the polarizability do not play an important role in our calculations. The reason is that in ⁴He the 2 ¹S₀ state (lifetime ≈ 20 ms) has a purely scalar polarizability and for the metastable 2 ³S₁ state (lifetime ≈ 7800 s) the vector and tensor polarizabilities are equal to zero within strict *LS* coupling.

The polarizability induced by an electromagnetic wave with angular frequency ω due to a single opposite parity state, in the LS coupling scheme, is [109, 146, 148]

$$\alpha^{(n)}(J, M_J, J', M'_J, q) = 6\pi\epsilon_0 c^3 (2J'+1) \begin{pmatrix} J & 1 & J' \\ -M_J & q & M'_J \end{pmatrix}^2 \frac{A_{nJJ'}}{\omega_{nJJ'}^2 (\omega_{nJJ'}^2 - \omega^2)}$$
(4.2)

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

$$\alpha(J, M_J, q) = \sum_n \sum_{J'} \alpha^{(n)}(J, M_J, J', M'_J, q)$$
(4.3)

We note here that in other work equation 4.3 is commonly further simplified by a summation over all M_J states and possible polarizations q (see, e.g. [146]). As our experimental work specifically concerns the spin-stretched $2 {}^{3}S_{1}$ ($M_J = +1$) state [21, 22], we only calculate the polarizability for the $M_J = +1$ state and assume linearly polarized light (q = 0). For ³He the calculations specifically concern the spin-stretched $2 {}^{3}S F = 3/2$ ($M_F = +3/2$) and $2 {}^{1}S F = 1/2$ ($M_F = +1/2$) states. The summation in equation 4.3 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 [149].

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 by introducing the effective quantum number

¹The original published version of the paper gives the expression for unpolarized light $\alpha(J, M_J) = \alpha_0 + \alpha_2 \frac{3M_J^2 - J(J+1)}{J(2J-1)}$ which is common in theoretical literature [109, 146, 148].

 n^* [150]:

$$n^* = n - \sum_{r=0}^{\infty} \frac{\delta_r}{n^{*r}},\tag{4.4}$$

where δ_r are fit parameters and the quantity $n - n^*$ is commonly referred to as the quantum defect. For both the singlet and triplet series, equation 4.4 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 [150].

Extrapolation of the Einstein A coefficients is more complicated as there is no relation such as equation 4.4 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 equation 4.3 and defining the polarizability density per upper state energy interval as

$$\frac{\Delta \alpha^{(n)}}{\Delta E} = \frac{2\alpha^{(n)}}{E_{n+1} - E_{n-1}},$$
(4.5)

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' given by the Rydberg formula $E_n(n^*) = -R_{\infty}/n^{*2}$. For ease of notation we have omitted all the dependent variables of $\alpha^{(n)}$ as defined in equation 4.2. 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

$$\frac{\Delta \alpha^{(n)}}{\Delta E} = \frac{\alpha^{(n)}}{R_{\infty}} \frac{(n^{*2} - 1)^2}{2n^*},$$
(4.6)

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

$$\frac{d\alpha^{(n)}}{dE} = \frac{6\pi\epsilon_0 c^3}{R_\infty} (2J'+1) \begin{pmatrix} J & 1 & J' \\ -M_J & q & M'_J \end{pmatrix}^2 \frac{C_{nJJ'}(n^*)}{\omega_{nJJ'}^2(\omega_{nJJ'}^2 - \omega^2)}, \quad (4.7)$$

where we define

$$C_{nJJ'}(n^*) \equiv \frac{A_{nJJ'}(n^{*2}-1)^2}{2n^*}.$$
(4.8)

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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. However, as the extrapolation is a function of energy, it can be extended beyond the ionization potential to calculate continuum contributions to the polarizability. This method 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 equation 4.7. The ionization limit serves as a natural choice as the energy at which we would switch between the discrete sum and the integration method. But even for large enough n we would already be making a negligible numerical error in varying the exact cutoff energy E_c at which we switch between these methods. Therefore we perform the calculation using the sum-over-states method to an arbitrary cutoff at $E_c = -R_{\infty}/n_{max}^{*2}$ and continue with an integration over the remaining states as

$$\alpha^{\text{cont}}(J, M_J) = \sum_{J'} \int_{E_c}^{\infty} \frac{d\alpha^{(n)}}{dE} dE, \qquad (4.9)$$

where E is the energy of the corresponding state. As we use a low-order polynomial fit of equation 4.8 to calculate $d\alpha^{(n)}/dE$, the integral of equation 4.9 can provide an analytical solution. The total polarizability is therefore easily calculated as a sum-over-states part and an analytical expression

$$\alpha(J, M_J) = \alpha^{\text{cont}} + \sum_{n=1}^{n=n_{max}} \sum_{J'} \alpha^{(n)}.$$
 (4.10)

4.3 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 4.4 where we compare our results to other calculations.

The numerical convergence of equation 4.10 is tested by varying n_{max} . We find that 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 equation 4.10 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. We use the *ab initio* calculations of the level energies and Einstein A coefficients for the $n \leq 10$ levels in helium [149]. The higher level energies are extrapolated using equation 4.4 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 [151] as used in other recent work [55]) 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 $C_{n,I,I'}(E)$. As mentioned before, no advanced methods are used to calculate transiton matrix elements to higher states or doubly excited states in the continuum. Instead, the heuristic approach we use is to choose an extrapolation function that is smooth, continuous and provides a convergent integral in equation 4.9. Therefore $C_{nJJ'}(E)$ is fit with a polynomial of maximum order 2. 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. This indeed shows that the absolute accuracy of our continuum contributions will be the limiting factor in the accuracy of the calculated polarizabilities. We chose to use a second order polynomial function 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 in section 4.4.

4.4 Results

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 (next subsection), the ac polarizabilities are given, 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. Finally, 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 [55].

Author (year)	Ref.	$2 {}^{1}S_{0}$	$2 \ {}^{3}S_{1}$
Crosby and Zorn (1977) [Experiment]	[152]	729(88)	301(20)
Ekstrom et al. (1995) [Experiment]	[153, 154]		322(6.8)
Chung and Hurst (1966)	[155]	801.95	315.63
Drake (1972)	[156]	800.2	315.608
Chung (1977)	[157]	801.10	315.63
Glover and Weinhold (1977)	[158]	803.31	316.24
Lamm and Szabo (1980)	[159]	790.8	318.7
Bishop and Pipin (1993)	[160]		315.631
Rérat <i>et al.</i> (1993)	[161]	803.25	
Chen (1995)	[147]	800.31	
Chen and Chung (1996) , B Spline	[162]		315.630
Chen and Chung (1996), Slater	[162]		315.611
Yan and Babb (1998)	[145]	800.31666	315.631468
Mitroy and Tang (2013) , hybrid	[55]		315.462
Mitroy and Tang (2013), CPM	[55]		316.020
This work		801.19	317.64

Table 4.1: Comparison of calculations and measurements of M_J -averaged dc polarizabilities of the 2 ${}^{1}S_0$ and 2 ${}^{3}S_1$ states in units of a_0^3 .

dc polarizabilities

An overview of previously calculated and measured dc polarizabilities for the 2 ${}^{1}S_{0}$ and 2 ${}^{3}S_{1}$ states of 4 He is given in Table 4.1 together with our results. For convenience we give the polarizabilities in atomic units a_{0}^{3} (a_{0} is the Bohr radius), but they can be converted to SI units through multiplication by $4\pi\epsilon_{0}a_{0}^{3} \approx 1.64877 \times 10^{-41} \text{ CV}^{-1}\text{m}^{2}$. Furthermore, the dc polarizabilities are calculated using the common convention of summing over all M_{J} states [146].

There is general agreement between our results and previously calculated dc polarizabilities, but comparison with the work of Yan and Babb [145], 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). This means that the calculated continuum contributions in equation 4.9 provide an overestimate of the continuum contributions. Furthermore, as the difference is smaller than our calculated continuum contribution (7.1 a_{0}^{3} and 3.6 a_{0}^{3} , respectively) and following the discussion in section 4.3, we can conclude that our absolute accuracy is indeed limited by the exact calculation of the continuum contributions.

Table 4.2: Calculated magic wavelengths λ_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 α 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 μ 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 section 2.3 for details on those calculations.

λ_m [nm]	$d\alpha/d\lambda$	$\alpha [a_0^3]$	Laser	Lifetime	Nearest
· · /// []	$[a_0^3/nm]$	00 [000]	power [W]	[s]	transition
318.611	-7.00×10^{4}	-809.2			
319.815	-4.40×10^3	189.3	0.7	3	$2 {}^{3}S_{1} \rightarrow 4 {}^{3}P_{0,1,2}$
321.409	$-5.38 imes 10^2$	55.3	2.3	6	$2 {}^{3}S_{1} \rightarrow 4 {}^{3}P_{0,1,2}$
323.587	-1.48×10^2	17.2	7.3	6	$2 {}^{3}S_{1} \rightarrow 4 {}^{3}P_{0,1,2}$
326.672	$-5.48 imes 10^1$	-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	$2 {}^{3}S_{1} \rightarrow 3 {}^{3}P_{0,1,2}$
1557.3	0.0	603.8	0.2	205	$2 {}^{3}S_{1} \rightarrow 2 {}^{3}P_{0,1,2}$

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 - 2.5 μ m and an overview of the magic wavelengths is shown in Table 4.2. We also give the slope of the differential polarizability, which is used to estimate the sensitivity of the determined magic wavelength due to the accuracy of the calculated polarizabilities. Table 4.2 furthermore provides the trapping beam power required to produce a trap depth of 5 μ K and the corresponding scattering lifetime (see section 2.3) to indicate the experimental feasibility of each magic wavelength.

The magic wavelengths in the range 318-327 nm, as shown in figure 4.1, are mainly due to the many resonances in the singlet series. 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 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 4.2, there are four more magic wavelengths. The magic wavelength at 411.863 nm, previously predicted with nm accuracy [141], is the only one in this region with a small yet positive $2 {}^{3}S_{1}$ polarizability (see inset in figure 4.2). There are no more magic wavelengths in the range



Figure 4.1: 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). 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 4.2.

420-2500 nm, which is shown in figure 4.3, and the polarizabilities converge to the dc polarizabilities for $\lambda > 2500$ nm.

The ac polarizability of the 2 ${}^{1}S_{0}$ state can be compared to previous polarizability calculations from dc to 506 nm [147]. Combined with the dc polarizability comparison and the tune-out wavelength result for the 2 ${}^{3}S_{1}$ state, we find that the accuracy of our calculations is limited by the exact calculation of the continuum contributions. We note that the absolute continuum contributions and the corresponding uncertainty are approximately 5 times larger for the magic wavelengths around 320 nm than for 412 nm, as the shorter wavelengths are closer to the 2 ${}^{1}S$ 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 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.

For experimental purposes the most promising magic wavelength is 319.815 nm, as the polarizability of the 2 ${}^{3}S_{1}$ state is large enough to provide sufficient trap



Figure 4.2: 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 4.2. The inset shows the wavelength region 411-415 nm, displaying the magic wavelength at 411.863 nm and the tune-out wavelength at 414.197 nm.

depth at reasonable laser powers while the estimated scattering lifetime is still acceptable.

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 [55], 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 $d\alpha/d\lambda = -1.9 a_{0}^{3}/\text{nm}$ at the zero crossing. We find this tuneout wavelength at 414.197 nm (see inset in figure 4.2), 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 2.23 a_{0}^{3} difference in the calculated absolute polarizabilities. Comparison of the calculated dc polarizabilities (see Table 4.1) 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.



Figure 4.3: 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 μ 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.

4.5 Extension to ³He

The 2 ${}^{3}S \rightarrow 2 {}^{1}S$ transition is also measured in ³He in order to determine the isotope shift of the transition frequency [21]. Hence a magic wavelength trap for ³He will be required as well. As ³He 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 ³He energy level data [30] and recalculating the quantum defects using equation 4.4. The Einstein A coefficients of the transitions also change due to the different reduced mass of the system [149], 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 [163]. 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 [164]. 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 npelectrons [163]. 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 ³He⁺ in the 1s ground state, there are two Rydberg series in the ³He atom. For even higher n than discussed before, this leads to mixing of Rydberg states with different n [163]. This leads to shifts that are well below the accuracy of the calculations and is 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 equation 4.2, but with substituted quantum numbers $(J, M_J \to F, M_F)$, Einstein A coefficients and transition frequencies. The numerical calculation of the polarizabilities and discussion of the numerical accuracies is similar to the ⁴He case. An additional uncertainty of 1.0 a_0^3 is added in the calculation of the polarizabilities of the ³He 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 ³He or ⁴He has a significant non-scalar polarizability for the states discussed in this paper.

A comparison between the ⁴He and ³He magic wavelengths is presented in Table 4.3. 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 4.3) 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 4.3 as they are not useful due to the large relative uncertainty. Table 4.3: Comparison of magic wavelengths λ_m calculated for the ⁴He 2 ³S₁ ($M_J = +1$) $\rightarrow 2$ ¹S₀ and ³He 2 ³S F = 3/2 ($M_F = +3/2$) $\rightarrow 2$ ¹S 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 ³He.

$\lambda_m \text{ [nm]}$		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	$-4(2) \times 10^{1}$

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.

4.6 Conclusion

We have calculated the dc and ac polarizabilities of the 2 ${}^{1}S$ and 2 ${}^{3}S$ states for both 4 He and 3 He in the wavelength range of 318 nm to 2.5 μ 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 paper 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 ³He. Although the hyperfine structure, which is absent in ⁴He, leads to complications in the calculation of the polarizabilities, these effects are very limited for the 2 ¹S and 2 ³S states. The magic wavelengths of interest, around 320 nm, are

shifted relative to the ${}^{4}\mathrm{He}$ magic wavelengths by predominantly the isotope shift.

4.A Appendix: Vector and tensor polarizability

The calculations in this chapter do not discuss the vector and tensor parts of the polarizability because both are expected to be small due to the high degree of LS coupling in the helium atom. Furthermore, linear polarization of the light is explicitly assumed in which case the vector part of the polarizability can safely be neglected. However, for the interpretation of the measurements in chapter 6 it becomes necessary to estimate the magnitude of the non-scalar part of the polarizability in order to verify that they do not influence the measurements. In this appendix a simple extension to the calculations is carried out to estimate the non-scalar terms in the polarizability.

In order to estimate the vector and tensor polarizabilities, I have extended the calculations of the polarizability in this chapter to include the $|2 \ ^{3}S_{1}; m_{J} = 0$ state for π -polarized light and of the $|2 \ ^{3}S_{1}; m_{J} = +1$ state for both σ^{+} and σ^{-} polarization. From these the vector and tensor polarizability, as defined by equation 4.1, are calculated according to

$$\alpha_1 = \alpha(m_J = +1, \sigma^-) - \alpha(m_J = +1, \sigma^+), \tag{4.11}$$

and

$$\alpha_2 = \frac{\alpha(m_J = +1, \pi) - \alpha(m_J = 0, \pi)}{3}.$$
(4.12)

At the magic wavelength the values $0.09 a_0^3$ and $0.03 a_0^3$ are found for the vector and tensor polarizabilities respectively. These numbers are small compared to the scalar polarizability of 189.3 a_0^3 , and the 0.5 a_0^3 uncertainty on the dc-offset on the polarizability curve as found in the magic wavelength measurement (see chapter 6).

Chapter 5

A laser system for trapping at the magic wavelength

Abstract

High precision spectroscopy on the 2 ${}^{3}S \rightarrow 2 {}^{1}S$ transition is possible in ultracold optically trapped helium but the accuracy is limited by the ac-Stark shift induced by the optical dipole trap. To overcome this problem, we have built a trapping laser system at the predicted magic wavelength of 319.8 nm. Our system is based on frequency conversion using commercially available components and produces over 2 W of power at this wavelength. With this system, we show trapping of ultracold atoms, both thermal (~ 0.2 μ K) and in a Bose-Einstein condensate, with a trap lifetime of several seconds, mainly limited by off-resonant scattering.

5.1 Introduction

The helium atom has proven to be a productive testing ground for fundamental physics. Frequency metrology has been employed as a sensitive test of QED calculations, both from the ground state [34, 35], and from the long lived (lifetime ~ 8000 s) metastable 2 ${}^{3}S$ state (He^{*}) [22, 39, 40]. Another interesting target for spectroscopy is to probe the influence on level energies of the finite size of the nucleus. By comparing accurate atomic structure calculations [32] to high precision isotope shift measurements, nuclear charge radii relative to the (accurately known [71]) ⁴He nucleus can be extracted. This method was employed to determine charge radii of the halo nuclei ⁶He and ⁸He [83] but also to measure the ⁴He-³He differential nuclear charge radius [21, 37]. These measurements are relevant to current investigations into the so-called "proton radius puzzle" which arose when a similar measurement of the proton radius in

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 μ H found a 7 σ discrepancy with the 2010 CODATA value [73]. Current efforts investigating the nuclear charge radii of μ^{3} He⁺ and μ^{4} He⁺ are projected to reach an experimental uncertainty at the sub-attometer (am) level [80]. Determinations of the ⁴He-³He differential nuclear charge radius with comparable accuracy in electronic systems provide a valuable cross-check for these measurements.

Currently the two most accurate measurements of the ⁴He-³He differential nuclear charge radius have achieved accuracies of 3 [37] and 11 am² [21], roughly an order of magnitude less precise than the projection of the μ He experiment, but disagree by 4σ . The former experiment resolved the $2 \ ^3S \rightarrow 2 \ ^3P$ transition to within one-thousandth of the 1.6 MHz natural linewidth and is not expected to be significantly¹ improved upon in the near future. The latter experiment was performed on the doubly forbidden $2 \ ^3S \rightarrow 2 \ ^1S$ transition whose 8 Hz natural linewidth is not a limiting factor but has a very low excitation rate and thus requires a long interaction time. To achieve this He^{*} atoms were cooled to quantum degeneracy (a Bose-Einstein condensate (BEC) of ⁴He^{*}, and in a degenerate Fermi gas of ³He^{*}) and trapped in an optical dipole trap (ODT). The accuracy of this experiment was limited by experimental effects, mainly the ac-Stark shift induced by the ODT.

This problem is also encountered in optical lattice clocks where it is solved by employing so-called magic wavelength traps [88]. In a magic wavelength trap, the wavelength of the trapping laser is chosen such that the upper and lower state polarizability are exactly the same, cancelling out the differential ac-Stark shift. In helium a high-precision calculation of the ac-polarizability of the 2 ${}^{3}S$ level was recently reported [55], and we ourselves have made more approximate calculations on both the 2 ${}^{3}S$ and 2 ${}^{1}S$ levels (see chapter 4). Both works predict the polarizability of the 2 ${}^{3}S$ level to vanish at around 413 nm (a so-called tune-out wavelength) which was later confirmed experimentally [56]. Our calculations also predict a number of magic wavelengths for the 2 ${}^{3}S \rightarrow$ 2 ${}^{1}S$ transition. The most promising from the perspective of trapping is located at 319.815 nm for ⁴He and 319.830 nm for ³He.

Trapping atoms at this wavelength is not straightforward. First of all, to achieve a trap depth comparable to [21], where an infrared ODT at 1557 nm was used, an optical power of approximately 1 W is required. Such powers are not readily available at ultraviolet wavelengths. Secondly, the lifetime of atoms trapped at this magic wavelength is intrinsically limited by two mechanisms: off-resonant excitation to the nearby $2^{3}S \rightarrow 4^{3}P$ transition at 318.9 nm,

¹After publication of this work a new measurement of this transition was reported [38] which improves the experimental accuracy by almost a factor two compared to the work referenced here but disagrees with it by 20σ .

and two-photon ionization. The total loss rate from these processes should not exceed about 1 s^{-1} , to allow sufficient probe time for spectroscopy.

Considerable progress has been made in the production of laser light in the wavelength range near 320 nm, primarily for the purpose of laser cooling Be⁺ ions [143, 144, 165–167]. High power (several hundreds mW) was generated by sum frequency mixing and subsequent frequency doubling of two fiber lasers, retaining most of their high spatial and spectral mode quality [143, 144, 167]. Production of up to 2 W was demonstrated with such a system [144]. Constrained to commercial Er and Yb doped fiber amplifiers, this scheme allows the production of high power continuous wave laser light over a range of 310-325 nm.

In section 5.2, we demonstrate a laser system built out of commercially available components producing over 2 W at 319.8 nm based on a modification of this scheme. In section 5.3 we show that our source can be used to trap helium atoms with an acceptable lifetime of a few seconds, such that spectroscopy on the 2 ${}^{3}S \rightarrow 2 {}^{1}S$ transition can be performed.

5.2 Laser system

The system can be divided in a sum frequency generation (SFG) part, which generates 639.6 nm light from two infrared lasers, and a second harmonic generation (SHG) part which frequency doubles the SFG light to 319.8 nm. In the following, we will first give a full overview of the optical system before discussing the results and performance of the SFG and SHG parts separately.

Overview

Figure 5.1 shows a schematic overview of the optical setup. The setup is relatively compact with all of the components, except for the lasers and the control electronics, mounted on a single $1000 \times 500 \text{ mm}^2$ optical breadboard. The system starts with two fiber lasers (NKT photonics Koheras Adjustik E15 and Y10) with center wavelengths of 1557.28 nm and 1085.45 nm. The thermal tuning ranges are 1000 pm and 700 pm respectively, covering a spectral range much larger than the uncertainty in the calculated magic wavelength. The lasers seed two 10 W fiber amplifiers (NuFern NUA-1084-PB-0010-C2 and NUA-1550-PB-0010-C2), with isolated polarization maintaining free-space output couplers. The beams are separately focussed to achieve optimal sum frequency generation.

The output beams are to an excellent degree of approximation Gaussian, and can be described completely by two parameters: their (minimum) waist size (w_0) , which is directly related to their Rayleigh range $(z_R = \pi w_0^2/\lambda)$,



Figure 5.1: Schematic view of the UV laser system. Two infrared fiber lasers at 1085.5 nm and 1557.3 nm seed two 10 W amplifiers. The infrared beams are independently focussed, and overlapped on a dichroic mirror (DM). The combined beam passes through a temperature stabilized PPLN crystal. More dichroic mirrors filter residual infrared light from the SFG beam at 639.6 nm. This beam is mode-matched and phase modulated by a 20 MHz electro-optic modulator (EOM) and the cavity reflection is monitored using a photodiode (PD) to allow Pound-Drever-Hall locking of the cavity. The final UV output beam is then collimated and ellipticity is compensated by an anamorphic prism pair (APP).

and the position of their focus. In order to achieve optimal conversion these parameters must be matched both to each other and to the crystal. Achieving this condition is not entirely straightforward because of the coupled nature of the problem.

We first collimate the 1557.3 nm beam with an f = 300 mm lens, and then focus by two lenses with focal distances of -100 mm and 200 mm. The beam waist can now be changed by moving either of the focussing lenses, but doing so will also move the focal point. The 1085.5 nm beam is first passed through a telescope consisting of two lenses with focal distances of 200 mm and 50 mm and is then focussed by an f = 300 mm lens. The focal point can be changed without affecting the waist size by moving the focussing lens. In this way, the foci of the beams are matched by first setting the waist of the 1085.5 nm beam to the desired focussing, secondly matching the 1557.3 nm beam waist to it, and finally overlapping the focal point of the 1085.5 nm beam with that of the 1557.3 nm beam.

With fixed beam parameters, the infrared beams are overlapped on a dichroic mirror and passed through a 40 mm MgO doped periodically poled lithium niobate (PPLN) crystal (Covesion) with a poling period of 12.1 μ m. The crystal is mounted in an oven and temperature stabilized at ~ 90°C. The output beam from the crystal contains both the sum frequency and residual infrared light. This residual light is filtered from the beam by two dichroic mirrors and the SFG beam is collimated by a f = 250 mm lens to a waist of ~ 1 mm.

The light is then coupled (free-space) into a commercial frequency doubling system (Toptica SHG pro) where it is mode-matched to the cavity and passed through an electro-optical modulator (EOM). The EOM modulates 20 MHz sidebands on the laser carrier frequency to allow Pound-Drever-Hall locking of the cavity. The doubling cavity is similar to [168], the main differences being the locking scheme (Pound-Drever-Hall instead of Hänsch-Couillaud) and the crystal (AR-coated rather than Brewster cut). The UV output is collimated and passed through an anamorphic prism pair to reduce beam ellipticity. Based on the specifications of the seed lasers and amplifiers the spectral linewidths of the infrared beams should be of the order of several tens of kHz. Because the nonlinear conversion steps do not significantly add to the fractional linewidth the final UV-output is expected to have a linewidth of ~ 100 kHz, which is small compared to the scale at which the polarizability changes [169].

Sum Frequency Generation

The purpose of the SFG stage is to convert the available infrared laser light into useful SFG light with high efficiency. To achieve this it is necessary to focus the input beams tightly so that a high peak intensity is reached, but not so tightly that the beams diverge too quickly before they reach the end of the crystal. As described by Boyd and Kleinman [170], this process can be optimized with respect to the dimensionless focussing parameter $\xi = l/2z_R$, which is the ratio of the crystal length l to the confocal parameter of the beam (twice the Rayleigh length z_R). Although optimal at $\xi \approx 2.84$, the efficiency varies quite slowly so that at confocal focussing ($\xi = 1$) it is still approximately 80% of its maximum value. The system produces more power than required and the confocal condition is chosen because of practical considerations such as the available path length and the size of the entrance surface of the crystal.

The infrared beams were set to the confocal condition with waist sizes measured to be 56(1) μ m ($z_R = 8.9(3)$ mm) for the 1085.5 nm beam and 63(1) μ m ($z_R = 7.9(3)$ mm) for the 1557.3 nm beam and their waist positions located within 1 mm of each other. By correcting for the refractive index of the crystals, which can be calculated based on known Sellmeier coefficients [171],



Figure 5.2: Results of sum frequency generation. (a) Sum frequency power as a function of the input power product (IPP) of the infrared lasers. The dashed line is a linear fit at low input powers (slope $0.108(1) \text{ W}^{-1}$). (b) Contourplot of the sum frequency power as a function of both input powers, based on a linear interpolation of the same dataset as (a). Black diamonds indicate measured datapoints, the dashed lines indicate contours of constant IPP.

the focussing parameter inside the crystal is found to be $\xi = 1.03$ for the 1085.5 nm beam and $\xi = 1.16$ for the 1557.3 nm beam.

Figure 5.2a shows the converted power as a function of the product of the input powers (input power product, IPP). When different combinations of input powers with equal IPP are used they produce almost the same SFG output power. This can be seen more clearly in a contour plot of the output power as a function of input powers (figure 5.2b, based on a linear interpolation of the same dataset) where contours of constant IPP follow constant output power. From this we conclude that the total converted power is a function of IPP only, and does not depend on the exact composition of infrared powers.

In order to achieve the power conversion plotted in Figs. 5.2a and 5.2b, it is necessary to optimize for crystal temperature at each input power product. The reason for this is shown in Figure 5.3, which shows the conversion efficiency as a function of crystal temperature at different input powers. As the input power becomes higher, the optimal temperature shifts to lower temperature and the crystal temperature needs to be adjusted to achieve maximum conversion. A possible explanation for this behaviour is that light is absorbed in the center of the crystal and heats it locally. This causes a slightly elevated temperature, and consequently imperfect phase matching at the center of the crystal (where conversion occurs) compared to the crystal edge (with respect to which the temperature is controlled). When the crystal temperature is set to a slightly lower temperature, this effect is compensated.



Figure 5.3: Sum frequency production as a function of crystal temperature, normalized for measured input power product. The plot shows input power products of 1.4 W² (blue circles), 35.7 W² (red squares), and 78.0 W² (black diamonds). At higher input powers the temperature graph becomes slanted towards lower temperature, indicating thermal effects.

At low input powers the SFG output power scales linearly with a slope of 0.108(1) W⁻¹, comparable to other experiments using a similar crystal [143, 144]. At higher output powers a deviation from the linear behaviour is seen. This may be a left-over thermal effect, or it may be that, because of the high conversion efficiency, pump depletion needs to be taken into account. In the case of a thermal effect the spatial output mode may be distorted but this is not observed. At an IPP of 80 W², consisting of 8 W at 1557.3 nm and 10 W at 1085.5 nm, a maximum output power of almost 6 W of SFG light is produced, which corresponds to a conversion efficiency of 33%.

Second Harmonic Generation

The generated SFG light is coupled in free space to the Toptica SHG pro system. Using a commercial frequency doubling system has advantages, but the disadvantage is that some system parameters are not disclosed. We will therefore describe the system as a whole with reported in- and output powers measured before and after the full system.

Figure 5.4 shows the system output power and conversion efficiency of the SHG section. At an input power of ~ 2 W the conversion efficiency saturates at about 50% and at an input power of 4 W a maximum output power of more than 2 W of UV light was achieved. At this point the cavity coupling efficiency is just over 80%. This behaviour is qualitatively similar to what is observed in other SHG systems [168, 172]. Although more SFG input power is available, we



Figure 5.4: Results from the SHG system. (a) UV output power (319.8 nm) as a function of total input power (639.6 nm) going into the full system. The output increases quadratically at low input powers, but saturates to a linear asymptote at higher powers. (b) System conversion efficiency (ratio of total input and output power) as a function of input power. The efficiency saturates at $\sim 50\%$.

choose to remain below 4 W input power to prevent (UV-induced) damage to the cavity optics at these powers [173]. Peak-to-peak UV intensity fluctuations of 4% were measured over a 4 hour period. This is within the specifications for intensity stability of the fiber amplifiers. No evidence of degradation of the crystal or cavity optics has been observed after more than a year of operation although periodic realignments of the cavity mirrors were required.

The beam profile of the cavity output is still Gaussian ($M^2 < 1.2$), but shows ellipticity due to crystal walk-off. Directly from the cavity we observe a beam waist of ~ 0.7 mm in the vertical and ~ 0.1 mm in the horizontal direction. This beam is passed through a collimating lens and an anamorphic prism pair to produce a more circular beam. We measure the final beam waist of ~ 0.21 × 0.35 mm². Despite the modest ellipticity these beam parameters still allow tight focussing, an essential requirement for optical dipole trapping.

5.3 Trapping

Now that we are able to produce sufficient power at 319.8 nm, we implement the laser system into our existing setup [21, 22] to demonstrate trapping and to characterize the trap lifetime. We prepare the beam for trapping by enlarging it with a 1:2 telescope and focus it inside the vacuum chamber with an f = 400 mm lens to a waist of $w_0^{(1)} \times w_0^{(2)} = 64.3(1.0) \times 55.6(7) \ \mu\text{m}^2$. The focus positions along the horizontal and vertical axes are found to lie 12(2) mm apart which is small compared to the Rayleigh lengths (40.0(6) mm and 30.0(4) mm). We measure 62% total transmission of the two windows of the vacuum chamber. While high, these losses are expected from uncoated sapphire vacuum windows [174].

A transmission per window of $T_1 = \sqrt{0.62} \approx 0.78$ is assumed to estimate the power inside the vacuum chamber. Therefore, at a power (P) of 1 W and neglecting astigmatism, the beam has peak intensity

$$I_p = \frac{2T_1 P}{\pi w_0^{(1)} w_0^{(2)}} = 14 \text{ kWcm}^{-2}.$$
 (5.1)

Based on the polarizability (calculated in chapter 4) this translates to a trap depth of ~ 6.0 $\mu K W^{-1}$ for a single beam. The trap depth of our ODT is therefore far below the recoil temperature

$$T_{rec} = \frac{\hbar^2 k^2}{k_B m} = 46.7 \ \mu \text{K}, \tag{5.2}$$

where $k = 2\pi/\lambda$ is the photon wavenumber and *m* is the atomic mass of helium. We can therefore safely assume that each photon scattering event leads to the loss of the scattered atom. Additionally, the excess energy of the scattered atom can heat the other atoms or even kick more atoms out of the trap.

When two- and three-body losses can be neglected, the total loss rate is a combination of three distinct rates: a background loss rate R_{bg} due to the background pressure inside the chamber, a loss rate R_{sc} due to photon scattering, and a loss rate R_{ion} due to two-photon ionization. These mechanisms scale in different ways with ODT power. The total loss rate is

$$R_{tot} = R_{bg} + R_{sc}I_p + R_{ion}I_p^2.$$
(5.3)

The Rayleigh length of the focussed UV beam is comparable to the ~ 4 cm spacing between the vacuum windows. Therefore, in a single beam ODT trapped atoms are able to collide with the windows and leave the trap. To avoid this some means of axial confinement is necessary. We use two different methods to provide this confinement. The first is to add a magnetic field gradient along the beam direction to create a hybrid trap [175]. With this method the loss rate and trap depth are straightforward to interpret. However, peak densities are not high enough to produce a BEC. The other method is to create a two-colour crossed-beam ODT using the UV beam and additionally a focussed 1557 nm beam which was already in place [21]. This trap gives a high enough peak density for a BEC to form.

The final magic wavelength trap for precision spectroscopy will be of a different geometry however, because both trapping schemes disussed here still introduce systematic shifts of the transition frequency. The most straightforward final trap geometry would be a crossed beam ODT using only the UV



Figure 5.5: Schematic view of the hybrid trap geometry. Atoms are trapped by the combination of the UV-laser beam and the quadrupole magnetic trap (QMT) coils. A set of fine-tune coils allows precise tuning of the trap center. Because of limited optical access the UV laser beam is at an angle of $\sim 9.5^{\circ}$ with the magnetic field axis. High-resolution detection is done using a micro-channel plate detector (MCP). Inset: in-situ absorption image of atoms in the UV hybrid trap.

laser. In such a trap the atomic density will be similar to what is found in the two-colour trap while the trap depth and scattering rate are comparable to the hybrid trap. Measuring these quantities therefore gives an accurate picture of what can be expected while requiring no modification of the current experimental setup.

Hybrid trap

We prepare an ultracold sample in a way previously described [21]. A beam of He^{*} atoms is generated from a liquid nitrogen cooled dc-discharge, collimated, slowed in a Zeeman slower, and captured in a magneto-optical trap. Here the atoms are cooled to approximately 0.5 mK. Subsequently they are spin-polarized, loaded into a Ioffe-Pritchard type magnetic trap and Doppler cooled to approximately 130 μ K. Finally, the atoms are cooled to ~ 0.2 μ K by forced rf evaporative cooling inside the magnetic trap.

The cloud is transferred to the hybrid trap consisting of the UV beam and a quadrupole magnetic trap (QMT) generated by a set of QMT coils in anti-Helmholtz configuration. Figure 5.5 shows a schematic of this trap. The quadrupole field has a strong axis gradient of 0.54 Gauss/cm but gravity acts in a direction perpendicular to this axis. In this direction the gradi-



Figure 5.6: Hybrid trap one-body loss rate as a function of UV power. The blue line is a linear fit with a slope of 0.16(2) s⁻¹W⁻¹, corresponding to off-resonant scattering.

ent is only half the magnitude which is well below the leviation gradient of $mg/\mu = 0.351$ Gauss/cm [175]. Below this gradient gravity is stronger than the confining magnetic force so that atoms are not trapped in the absence of the UV beam. The QMT therefore adds confinement but does not contribute to the trap depth. A homogeneous magnetic field is applied with a set of fine-tune coils to minimize trap oscillations induced by the loading step.

The atoms are detected either by absorption imaging or by a micro-channel plate detector (MCP) located 17 cm below the trap center. MCP time of flight measurements are done as a function of hold time in the hybrid trap. The time of flight signals are fitted with a thermal Bose-Einstein distribution to extract the temperature of the gas as well as the atom number. The decay in atom number is fitted with an exponential (with an oscillating component to account for residual trap oscillations). The first few seconds of the decay are not fitted to neglect two-body loss and thermalization effects.

Figure 5.6 shows the loss rate of a thermal gas inside the hybrid trap. The loss rate varies linearly with power, which is consistent with only background collisions and off-resonant scattering; two-photon ionization would depend quadratically on power. A linear fit gives a slope of about $0.16(2) \text{ s}^{-1} \text{W}^{-1}$ ($R_{sc} \approx 1.2 \text{ s}^{-1} \text{W}^{-1} \text{m}^2$, see equation 5.3), with a background loss rate of $0.12(2) \text{ s}^{-1}$. This background loss rate is consistent with the loss rate of $0.13(1) \text{ s}^{-1}$ in a hybrid trap of comparable depth using our 1557 nm ODT for which off-resonant scattering is negligible [169].

Figure 5.7 shows the fitted temperature as a function of hold time in the hybrid trap. This temperature is not constant; after a quick thermalization the temperature starts to increase linearly with time, however, not fast enough



Figure 5.7: (a) Temperature as a function of hold time in the hybrid trap. After some thermalization time the atomic cloud starts to heat as a result of UV absorption. The blue dotted line is a constant fit to the temperature around the minimum to extract the minimum temperature. The red dashed line is a linear fit to the temperature after a few seconds to extract the heating rate. (b) Determination of the trap temperature based on minimum temperature (blue circles), and an extrapolation to zero hold time of the heating rate (red squares). From linear fits to these data an upper (lower) bound is set on the temperature in absence of heating of $0.62(2) \ \mu K \ W^{-1} (0.51(2) \ \mu K \ W^{-1})$.

to pose a problem. Two generic sources of heating in a dipole trap are intensity noise and beam pointing noise [176]. The former is not observed at this timescale because it would lead to exponential rather than linear heating. Beam pointing noise is a possible explanation of the observed heating but was never observed to be a problem in our infrared trap which uses a similar geometry. A more plausible heating mechanism is that atoms off-resonantly scattering a photon dump a small portion of their high recoil energy in the atomic cloud, thereby heating the ensemble. In principle it is also possible that a fraction of atoms heated by scattering is able to dump all recoil energy in the cloud and thermalize instead of leaving the trap. This may cause the photon scattering rate that was determined earlier to be an underestimate because a part of all scattering events appear as heating rather than trap loss. To assess the maximum contribution to the scattering rate of this effect we assume the most extreme case in which the rethermalization of recoiling atoms causes all of the observed heating. In this case, the highest observed heating rate of $\dot{T} \approx 0.04 \ \mu \text{K s}^{-1}$ corresponds to no more than $\dot{T}/T_{rec} \approx 0.001 \ \text{s}^{-1}$ unaccounted scattering events. This is two orders of magnitude lower than typically observed trap loss and can be safely disregarded.

To give an estimate of the equilibrium temperature inside the trap (in the absence of heating) we take the minimum achieved temperature as an upper bound, and a linear extrapolation of the heating to zero hold time as a lower bound. In this way we extract temperatures of $0.62(2) \ \mu \text{K W}^{-1}$ and $0.51(2) \ \mu \text{K W}^{-1}$ respectively. This is approximately a factor 10 lower than the



Figure 5.8: (a) Schematic picture of the two-colour ODT setup. The angle between the beams is 19°. (b) MCP time-of-flight measurement of a BEC in a two colour optical dipole trap. The signal is fit with a bimodal distribution indicating a BEC (red dashed line) and a thermal fraction (blue dotted line).

calculated trap depth and corresponds to a truncation parameter $\eta = 10$ which is typically found in a thermalized trapped gas [175].

Two-colour trap

Because of the low confinement provided by the hybrid trap, no BEC was observed. To provide enough confinement to observe BEC we switch to a two-colour trap consisting of the UV beam and an IR beam (focussed to a waist of 85 μ m) which cross at an angle of 19°. Figure 5.8a shows a schematic of the setup. Figure 5.8b shows the corresponding time-of flight signal on the MCP, fit with a bimodal distribution. Superposed on the thermal distribution is a clear inverted parabola Thomas-Fermi profile demonstrating Bose-Einstein condensation. For this BEC a one-body lifetime is observed of ~ 4s.

5.4 Conclusion

In order to perform magic wavelength trapping of metastable helium atoms we have realized a laser system which produces over 2 W at 319.8 nm, with not yet an indication of a reduction of SHG efficiency at higher pump power. The setup is built from commercially available fiber lasers, amplifiers and SFG/SHG components. Similar performance should be possible in a spectral range of 310-325 nm with only minor changes in the required components (mainly limited by available wavelength ranges of the amplifiers). The produced UV light is used to trap an ultracold (~ 0.2 μ K) thermal gas in a hybrid trap and a BEC in a two colour ODT. Trap losses are found to be mainly due to off-resonant scattering with a rate of 0.16(2) s⁻¹ W⁻¹.

With this system we can make a sufficiently deep dipole trap in the UV while keeping the intrinsic losses at an acceptable level such that spectroscopy is possible. This opens the door to a full magic wavelength ODT and a more precise measurement of the $2 {}^{3}S \rightarrow 2 {}^{1}S$ transition frequency.

Chapter 6

Precision spectroscopy in a magic wavelength trap

Abstract

Improvements in both theory and frequency metrology of few-electron systems such as hydrogen and helium have enabled increasingly sensitive tests of quantum electrodynamics (QED), as well as ever more accurate determinations of fundamental constants and the size of the nucleus. At the same time advances in cooling and trapping of neutral atoms have revolutionized the development of increasingly accurate atomic clocks. Here, we combine these fields to reach the highest precision on an optical transition in the helium atom to date by employing a Bose-Einstein condensate confined in a magic wavelength optical dipole trap. The measured transition accurately connects the ortho- and parastates of helium and constitutes a stringent test of QED theory. In addition we test polarizability calculations and ultracold scattering properties of the helium atom. Finally, our measurement probes the size of the nucleus at a level exceeding the projected accuracy of muonic helium measurements currently being performed in the context of the proton radius puzzle.

6.1 Introduction

In the past decades, high-precision spectroscopy measurements in atomic physics scale systems have pushed precision tests of quantum electrodynamics (QED), one of the cornerstones of the standard model of physics, ever further [16, 31] and have led to accurate determinations of fundamental constants [51, 65, 66, 177]. Recently however, measurements of transition frequen-

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cies in muonic hydrogen (μ H) have revealed a discrepancy of six standard deviations [72, 73] with respect to the accepted CODATA value for the proton charge radius. This discrepancy, which has become known as the "proton radius puzzle", has stimulated strong interest in the field, as its confirmation implies the violation of lepton universality, one of the pillars of the standard model. New experiments in atomic hydrogen [17, 18], and muonic deuterium [74] have only deepened the puzzle, prompting research into other elements such as muonic helium (μ ^{3,4}He⁺) [80]. From these measurements the charge radii of the alphaparticle and the helion (1.68 fm resp. 1.97 fm) are projected to be determined with sub-attometer accuracy, which should be compared to high-precision experiments in electronic helium atoms or ions.

QED theory of the helium atom, with two electrons more complicated than hydrogen, has seen impressive improvements in recent years, with QED corrections up to order $m\alpha^6$ now evaluated [31]. Recent experiments are in good agreement [22, 34, 36, 38, 39, 41, 42, 52] and may allow a competitive value for the fine structure constant in the near future [45, 46, 48, 49]. The anticipated evaluation of the next highest order corrections $(m\alpha^7)$ [31] would allow the determination of the ⁴He nuclear charge radius with an accuracy better than 1%. At present nuclear charge radii can already be determined differentially, i.e. with respect to ⁴He, due to cancellation of higher-order terms in the isotope shift. Using this approach the radii of the exotic halo nuclei ⁶He and ⁸He [82, 83], as well as the stable isotope ³He [21, 37, 84] were determined with accuracies far exceeding electron scattering experiments [81]. However, different experiments on the ³He-⁴He isotope shift show significant discrepancies [31], even between different measurements of the same dipole allowed $2 {}^{3}S_{1} \rightarrow 2 {}^{3}P$ transition [38]. Furthermore, improving the experimental accuracy on this transition is challenging due to the 1.6 MHz natural linewidth and the presence of quantum interference shifts [49]. Only one previous experiment has used the doubly forbidden $2 {}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition [21], which in contrast has an excellent quality factor of 2.4×10^{13} (natural linewidth 8 Hz) that poses no fundamental limit in the foreseeable future.

Here we report a new measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency at 1557 nm which improves the previous result by an order of magnitude, making this the most accurate optical frequency measurement in the helium atom to date ($\delta\nu/\nu = 1.0 \times 10^{-12}$). Our measurement has been performed using a Bose-Einstein condensate (BEC) in the metastable 2 ${}^{3}S_{1}$ state confined in an optical dipole trap (ODT) at a previously predicted [169] magic wavelength for this transition. At such a magic wavelength the ac-Stark shift on the transition vanishes, a property that has been exploited to realize atomic clocks operating at a stability in the 10^{-19} region [19, 20], allowing constraints on a possible time-variation of fundamental constants [142]. Moreover, ab-initio calculations of polarizability have recently emerged as an alternative means of testing atomic theory at a level sensitive to QED effects [55, 56, 178]. By finetuning the ODT laser wavelength to reduce the ac-Stark shift on the transition frequency, we measure the magic wavelength to high accuracy, providing a stringent test for ab-initio calculations [179].

Our approach has also enabled us to measure the mean-field, or coldcollision, shift on the transition for the first time by direct observation. This frequency shift was instrumental in the first observation of Bose-Einstein condensation of atomic hydrogen via two-photon excitation of the $1S \rightarrow 2S$ transition, where the enormous density of the BEC gave rise to a huge mean-field shift [15, 128]. The associated transition lineshape allowed quantitative analysis of these results [129]. In earlier work [63], we showed how this lineshape is affected by an asymmetry in the trapping potential for 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ atoms, and we could extract the 2 ${}^{1}S_{0} - 2 {}^{3}S_{1}$ scattering length with 50% accuracy. Now, working in a magic wavelength trap, we are able to improve this accuracy by an order of magnitude.

These measurements therefore test our knowledge of the helium atom in three different ways. The transition frequency measured here is a test of level energies and is sensitive to the finite size of the nucleus. The magic wavelength determination is a precision test of atomic structure as a whole and is therefore also sensitive to transition dipole moments. Finally, the scattering length derived from the mean-field shift is a precise test of the molecular potentials between helium atoms.

6.2 Setup

We prepare a BEC of typically 10^6 atoms in the metastable 2 3S_1 state (19.82 eV above the 1 ${}^{1}S_{0}$ ground state, lifetime ~ 8000 s [52]) [21], and transfer it into a dipole trap at 319.8 nm. The atoms are spin-polarized in the spin-stretched $m_{J} = +1$ state so that ionization via two-body collisions (Penning ionization) is strongly suppressed [7]. Figure 6.1a shows the geometry of the dipole trap. A tightly focused ODT beam is passed through the vacuum chamber, refocused and passed through the chamber again with orthogonal linear polarization, intersecting itself at an angle of 19°. The atoms are trapped at the intersection, where the probe laser is applied counterpropagating to the incident ODT beam. To detect excitation of the transition, we measure the increased Penning ionization rate from the excited 2 ${}^{1}S_{0}$ atoms using a microchannel plate detector (MCP) and counter (see section 6.5). This detection method provides substantially better signal-to-noise ratio compared to a signal based on the loss of 2 ${}^{3}S_{1}$ atoms used previously [21, 22, 63]. After excitation, the remaining atoms (>90%) are dropped under gravity on another MCP detector placed 17 cm directly below the trap, producing a time-of-flight (TOF)



Figure 6.1: a) Schematic of the experimental geometry. Two overlapping laser beams crossing at an angle of 19° form the ODT. The probe light is counterpropagating with one of the ODT beams. A high-voltage biased MCP detector above the setup detects ions generated by excited atoms. A grounded MCP below the setup detects the remaining metastable atoms that fall under gravity when they are released from the trap. b) Schematic of the laser setup. An erbium fiber laser (EFL) is transfer-locked in a phase locked loop (PLL) to an ultrastable erbium fiber laser (UEFL) via an optical frequency comb (OFC). Control over the frequency offset is provided by an in-loop direct digital synthesizer (DDS). The EFL serves as the probe laser, but part of it is also split off to seed a fiber amplifier. An independent ytterbium fiber laser (YFL) is amplified and overlapped with this light in order to generate the sum frequency (SFG), which is frequency doubled in a second harmonic generation (SHG) stage. A wavemeter (WM) is used to measure the wavelength of the YFL.

signal. From a bimodal fit to the TOF signal, we determine the chemical potential and atom number of the BEC, as well as the temperature and atom number of the thermal cloud.

Figure 6.1b shows the optical setup generating the probe and trap laser light. Part of the probe laser light is also amplified and mixed with a second independent laser in order to generate the ODT light. This second laser is monitored by a high resolution wavemeter to determine the trap laser wave-



Figure 6.2: A typical spectroscopy scan. The black circles indicate the signal when spectroscopy light is applied. The grey triangles are background calibration points measured directly after each spectroscopy point. In order to account for the Zeeman shift, the atoms are alternately excited from the $m_J = +1$, and $m_J = -1$ states. The red line is a fit of two Gaussians showing typical widths of about 10 kHz.

length (see section 6.5). The optical and electronic setup for generating the probe and ODT laser light are described in refs. [63] and [180].

To account for the Zeeman shift arising from the ambient magnetic field in the laboratory, we alternate between exciting from the $m_J = +1$ and $m_J = -1$ state (see section 6.5), which have first-order Zeeman shifts of equal magnitude but opposite sign. Exciting from the $m_J = 0$ state, which shows no firstorder Zeeman shift, is not possible due to a high Penning ionization rate [7]. Every measurement is alternated with a background measurement in order to monitor the level of background ion counts. In this way, we build up a doublepeak spectrum as shown in figure 6.2. We fit each measured line with two Gaussian peaks (see section 6.5) and calculate the center frequencies.

6.3 Results

By employing a magic wavelength ODT, the ac-Stark shift induced by the trap is greatly reduced compared to previous work [21]. The magic wavelength was not known with sufficient accuracy to eliminate the ac-Stark shift completely, and a residual trap-induced ac-Stark shift remains as a systematic shift that needs to be calibrated. In addition to this, two other systematic shifts are present that contribute roughly equally to the final accuracy: the ac-Stark



Figure 6.3: a-d) Results of a multiple regression fit to a single dataset. The graphs a) through c) are the partial residual plots for each of the fit parameters. The final graph d) shows the measured frequencies (blue open circles) and the residuals of the regression model (red circles). The grey band in all these figures indicates the 1σ uncertainty on the transition frequency determined from this particular dataset. e) Measured transition frequency per measurement week including systematic errors. The data point at week six is derived from the multiple regression fit shown in a-d, and the other points are based on similar datasets. The blue line and blue band indicate the weighted average and 1σ uncertainty.

shift from the probe laser, and the mean-field shift which is proportional to the chemical potential of the BEC.

In order to account for these systematics we performed multiple measurements in which we varied the ODT and probe laser powers as well as the chemical potential of the BEC. Since all of these systematic shifts are linear with respect to their corresponding experimental observable, we can fit the data with a multiple linear regression model, as shown in figure 6.3. From this model we extracted the transition frequency as well as the slopes of the ac-Stark shifts and the mean-field shift simultaneously. For every measurement week, a single complete fit of the regression model was performed, where the total number of measured transition frequencies varied between 16 and 39. It was experimentally not possible to vary all parameters independently. In particular the trap power and chemical potential are highly correlated because a deeper trap is better able to hold a high number of atoms at high density. To break this correlation as much as possible, we varied the chemical potential of the BEC while keeping the trap power fixed. This was achieved by varying the hold time in the ODT before applying probe light between 200 ms and a few seconds. Due to the fairly short (~ 2 seconds, limited by off-resonant scattering of the ODT light) one-body lifetime of the BEC in the ODT, this allows for significant modification of the size of the BEC.



Figure 6.4: a) Transition frequency as a function of laser power, showing only the "Coarse" scan, with a linear fit at each wavelength in order to determine the ac-Stark shift. b) Slopes of the fitted lines in a) as a function of ODT laser wavelength. The gray region indicates the predicted range for the magic wavelength [169]. c) "Fine" scan of the ac-Stark shift with the polarizability curve from ref. [169] fitted (red line). The blue region represents the 1σ uncertainty on the fit. The magic wavelength condition is found at the zero-crossing.

Magic wavelength

The determination of the magic wavelength was performed over two measurement campaigns: a first "coarse" campaign, and a second "high resolution" campaign during which the absolute transition frequency was also measured. The results from the coarse campaign are shown in figs. 6.4a,b, along with the predicted range from calculations [169]. These measurements gave a first estimate of the magic wavelength. However, during this first campaign the mean-field shift was not corrected for, leading to a small systematic offset on the magic wavelength.

During the final measurement campaign, the trap laser wavelength was varied over a smaller range in order to more precisely pinpoint the magic wavelength. Figure 6.4c shows the results from this campaign. In order to determine the magic wavelength we compare these data to the calculated polarizability curves [169]. The dominant uncertainty in these calculations depends only very weakly on wavelength and appears as a constant offset. The calculated polarizability can be related directly to the measured Stark shift by a scaling factor which corrects for the laser intensity (see section 6.5). By fitting the data to the calculated polarizability with a constant offset and a scaling factor, we also account for the slight curvature of the polarizability curve over the measured wavelength range. From this fit we then extract the laser intensity, a constant offset correction to the calculated polarizability, and the zero-crossing. We find the light intensity at the center of the trap to be $1.0(1) \times 10^8 \text{ Wm}^{-2}$ using a 1 W ODT beam. This intensity is roughly half of our estimate assuming perfect focusing conditions and beam quality [180]. The constant offset correction to the polarizability is found to be 3.4(5) a_0^3 . The zero crossing of the fitted curve corresponds to the magic wavelength and is found at 319.815 92(15) nm, which is in good agreement with the calculated value of 319.815(3) nm. The vector and tensor part of the polarizability are negligible at the current level of uncertainty (see appendix 6.A), and also do not influence the measurement of the transition frequency.

Mean-field shift and scattering length

The excited 2 ${}^{1}S_{0}$ atoms experience a different mean-field potential compared to the remaining 2 ${}^{3}S_{1}$ atoms because of the difference in scattering length. This leads to a shift of the transition frequency known as the meanfield, or cold-collision shift [128]. The full-width of the mean-field lineshape $S(\nu)$ [129] turns out to be small compared to the observed linewidth. At the maximum density used in the experiments (peak density $n(0) \approx 4.5 \times 10^{13} \text{ cm}^{-3}$ or equivalently $\mu \approx k_{B} \times 0.5 \ \mu\text{K}$), a full width $\delta \nu_{max} \approx 4.4 \text{ kHz}$ is expected (see appendix 6.A). The additional ac-Stark shift contribution to the width [63] is negligible for the range of ODT laser wavelengths used in the final measurement campaign. Possible line-pulling effects due to the asymmetric lineshape were investigated by fitting Gaussians to simulations of the broadened lineshape but were not found to affect the fitted frequencies.

The only observable effect of the mean-field interaction is therefore the average shift of this lineshape. We derive this average shift analytically by integrating the shift over the lineshape (see appendix 6.A for a detailed derivation):

$$\left\langle \Delta \nu_{MFS} \right\rangle = \frac{\int \nu S(\nu) d\nu}{\int S(\nu) d\nu} = \frac{4}{7h} \left(\frac{a_{tt} - a_{ts}}{a_{tt}} \right) \mu, \tag{6.1}$$

where a_{tt} and a_{ts} are the scattering lengths for triplet-triplet and triplet-singlet collisions respectively, and μ is the chemical potential of the BEC.

The mean-field shift slope was found by including a linear regression to the chemical potential of the BEC in the multiple regression model shown in figure 6.3c. Averaging over all measurements, we find a slope of -5.0(4) kHz μ K⁻¹. By rewriting equation 6.1 we can express the unknown triplet-singlet scattering length in units of the very well known triplet-triplet scattering length, $a_{tt} = +7.512(5) \text{ nm} = +142.0(1) a_0$ [57]. We find $a_{ts} = +82.5(5.2) a_0$, which is in agreement with our previous result of $a_{ts} = +50(10)_{stat}(43)_{syst} a_0$ [63].

Transition frequency

The final measured transition frequency is corrected for a number of systematic shifts as shown in table 6.1. By far the largest of these is the recoil shift correction due to the absorption of a 1557 nm photon, $\Delta f_{rec} = -h/(2m\lambda^2) = -20.554$ kHz, with negligible uncertainty.

Another systematic effect affecting all measurements equally is the frequency offset of the Cesium clock with respect to the SI-second to which all measurements are referenced. By comparing the clock with GPS time over the course of the entire measurement campaign, a fractional frequency offset of $-1.9(2) \times 10^{-13}$ was found (see section 6.5). By correcting for this offset, the clock was calibrated to within its specified stability floor of 5×10^{-14} , which contributes to the error budget. The measured transition frequency was corrected for the clock offset, corresponding to -36 Hz on the optical frequency.

Additional systematics are the black-body radiation shift, a dc-Stark shift due to the ion-MCP bias voltage, possible shifts due to quantum interference with far-off resonant transitions [49], and the second-order Zeeman shift. None of these contribute significantly to the final error budget, and could be neglected in the final result. Details of these estimations can be found in appendix 6.A.

Figure 6.3e shows the weekly average of all frequency measurements corrected for the systematic effects identified. Averaging over all results, we find Table 6.1: Measured 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency along with corrections. The final result is compared to several alternative determinations. Values are in kHz.

Term	Correction	Uncertainty
Measured frequency	$192\ 510\ 702\ 169.31$	
Recoil shift	-20.554	
ac-Stark shift: Probe)
ac-Stark shift: ODT		0.192^*
Mean-field shift		J
Statistical		0.032
Cs clock	-0.036	0.010
Black-body radiation shift	< 0.005	
dc-Stark shift	< 0.001	
Quantum interference	$< 10^{-4}$	
Second-order Zeeman	$< 10^{-5}$	
Total:	$192\ 510\ 702\ 148.72$	0.20
van Rooij <i>et al.</i> [21]	$192\ 510\ 702\ 145.6$	1.8
$IE(2 \ {}^{3}S_{1}) - IE(2 \ {}^{1}S_{0}) [30, 38, 41, 42]$	$192 \ 510 \ 702 \ 156$	42
Pachucki et al. [31] (theory)	$192\ 510\ 703\ 400$	800

* Uncertainty is correlated in the multiple regression model.

a 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency of 192 510 702 148.72(20) kHz, which corresponds to a relative uncertainty $\delta \nu / \nu = 1.0 \times 10^{-12}$.

6.4 Discussion and Conclusion

The magic wavelength found in this work is in very good agreement with our earlier calculation [169] but is more accurate by over an order of magnitude. Very recent full-configuration-interaction calculations incorporating relativistic and recoil effects give the magic wavelength as 319.816 07(9) nm, which is of a similar accuracy as our measurement and in excellent agreement [179]. It is interesting to make the comparison with measurements on the tune-out wavelength (the wavelength for which the polarizability vanishes) for the 2 ${}^{3}S_{1}$ level at 413 nm [56]. Here a discrepancy with high precision calculations was found which was attributed to QED effects [178], indicating that measurements of atomic polarizability can be used as an alternative means of testing QED.

The triplet-singlet scattering length a_{ts} derived from the mean-field shift measured in this work is more accurate than the previous experimental bound [63] by an order of magnitude, and in good agreement. This value can be used to test quantum chemistry calculations of the relevant molecular potentials. Interestingly, a previously reported estimate, derived from ab-initio calculations of the 1 ${}^{3}\Sigma_{g}^{+}$ and 2 ${}^{3}\Sigma_{g}^{+}$ molecular potentials [59], found $a_{ts} = +42^{+0.5}_{-2.5} a_{0}$ [63], which disagrees significantly with the value of $a_{st} = +82.5(5.2) a_{0}$ found in this work. This discrepancy may be related to the high ionization cross section which causes the complex optical potential method used in these calculations to break down.

The 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency obtained in this work improves the earlier measurement by van Rooij *et al.* [21] by an order of magnitude. The results differ by 1.7 σ (see table 6.1). This difference may be due to a slight underestimation of the mean-field shift in that work, which was reported as negligible at the level of 1.1 kHz. Based on the slope of the mean-field shift found in this work and a rough estimation of the chemical potentials used in ref. [21], we estimate that the mean-field shift in that work may have been somewhat larger (up to 2 kHz), which brings the results to within 1 σ of each other.

We can test for consistency with other experiments by taking the difference in ionization energy (IE) between the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ levels. These IEs can be determined from transition frequency measurements [38, 41, 42] and the theoretical IE of the 3 ${}^{1,3}D$ levels [30] which can be calculated to high accuracy. As shown in table 6.1, the measured transition frequency is in excellent agreement with this difference though more accurate by more than two orders of magnitude. The measured transition frequency is also in reasonable agreement (1.6 σ) with direct QED calculation [31], although the estimated uncertainty in this calculation is several orders of magnitude larger (see table 6.1). This uncertainty is currently of the same order as the total nuclear size shift, but is anticipated to be reduced [31] which would allow a direct determination of the ⁴He nuclear charge radius from the measured transition frequency.

At the current state of the theory, nuclear size information can still be derived at high accuracy by looking at the ³He-⁴He isotope shift on this transition, for which the estimated uncertainty in the calculations is much smaller (0.19 kHz) [31]. Taking the difference between the transition frequency measured in this work and the 2 ${}^{3}S_{1,F=3/2} \rightarrow 2 {}^{1}S_{0,F=1/2}$ transition frequency in ³He [21], we derive an updated value of the differential nuclear charge radius $\delta r^{2} = r^{2}({}^{3}\text{He}) - r^{2}({}^{4}\text{He}) = 1.041(7) \text{ fm}^{2}$ (see ref. [31] for details of the calculation), where the error is now dominated by the 1.5 kHz accuracy on the ³He transition frequency. This new value agrees with electron scattering $(\delta r^{2} = 1.066 \pm 0.06 \text{ fm}^{2} \text{ [81]})$ but still disagrees with determinations based on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{0,1,2}$ transition frequency in ⁴He [38] showed a 20 σ discrepancy with ref. [37], which also indicates the need for further investigation of that transition. In the immediate future, we aim to improve the measurement of the 2 ${}^{3}S_{1,F=3/2} \rightarrow 2 {}^{1}S_{0,F=1/2}$ transition in ³He, which may bring the uncertainty on δr^{2} down to $< 0.002 \text{ fm}^{2}$. This is actually better than the expected

accuracy from muonic helium, which will be limited to 0.0031 fm^2 because of theoretical uncertainty in calculating the two-photon exchange contribution to the Lamb shift [33].

The measurements presented in this work push our knowledge of the helium atom at several levels. The 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency is measured to be 192 510 702 148.72(20) kHz, intimately tying the ortho- and parastates together and allowing us to extract the ${}^{3}\text{He} - {}^{4}\text{He}$ nuclear charge radius difference with improved accuracy. The magic wavelength on this transition is determined to be 319.815 92(15) nm, in good agreement with calculations and provides a stringent test for precision calculations of polarizabilities. Finally, the measurement of the mean-field shift allows extraction of the 2 ${}^{3}S_{1} - 2 {}^{1}S_{0}$ scattering length as $+82.5(5.2) a_{0}$, which disagrees significantly with recent quantum-chemistry calculations.

6.5 Methods

Experimental sequence

The tightly focused (waist < 100 μ m) ODT beams trap the atoms in a cigar-shaped harmonic potential. At 1 W input power, typical trap frequencies are $\omega_{ax} \approx 2\pi \times 35$ Hz in the axial direction, and $\omega_{rad} \approx 2\pi \times 300$ Hz in the radial direction. The probe laser beam has an input power of up to 80 mW and a larger beam waist (~ 300 μ m) in order to ensure uniform illumination. We align the probe beam by overlapping it with the incident ODT beam. The polarization of the probe beam is linear but the direction is rotated with a motorized rotation stage in order to optimize the ion signal depending on whether the transition is made from the $m_J = +1$ or the $m_J = -1$ state.

In the ODT, probe light is applied for about 100 ms, after which the remaining atoms are released to fall under gravity onto the MCP detector. This excitation time is chosen to yield sufficient signal while being short enough to not alter the chemical potential of the BEC by more than a few percent. During this step, the excited 2 ${}^{1}S_{0}$ atoms collide with the remaining 2 ${}^{3}S_{1}$ atoms in a strongly Penning ionizing collision channel. We expect an ionization rate comparable to that of unpolarized 2 ${}^{3}S_{1}$ atoms, corresponding to a lifetime of about 1 ms for the 2 ${}^{1}S_{0}$ atoms. The He⁺ ions produced by this process are detected by a second MCP detector (the ion-MCP) biased at -2.5 kV and located 11 cm above the trap. The signal from the ion-MCP is amplified by a pulse amplifier/discriminator and passed into a counter to yield the spectroscopy signal. Based on an excitation fraction of ~ 5%, and a peak signal height of a few thousand counts from a BEC of a few million atoms, we estimate a detection efficiency of $\sim 2\%$. We attribute the low efficiency to shielding of the trap volume by the grounded re-entrant windows.

To mitigate the Zeeman shift, we alternate between exciting from the $m_J = +1$ and $m_J = -1$ state. We transfer the atoms from the $m_J = +1$ to the $m_J = -1$ state via a Landau-Zener sweep which consists of a magnetic field ramp while RF-coupling between the magnetic substates is applied [64]. After application of the probe light a second sweep brings the $m_J = -1$ atoms back to the m = +1 state so that the TOF is not affected. In the case of $m_J = +1$ atoms the same sweep is performed without the RF-coupling to make sure no systematic magnetic field difference is introduced. After every measurement an identical measurement is performed with the probe light blocked in order to calibrate the level of background ion counts. We attribute this background to ionization of background gas by the 320 nm ODT light. This is corroborated by the fact that the background count rate is linearly proportional to the ODT laser power and increases when the background pressure is increased by temporarily closing the safety valve going to the main chambers' turbopump.

We estimate the linewidth of the probe laser at about 5 kHz, based on the combined effects of residual frequency comb noise, electronic noise on the phase-locked loop, and the 60 meter uncompensated fiber link between the frequency comb and the setup. The observed lineshapes are broader however, showing approximately Gaussian profiles of about 10 kHz width. We attribute the additional broadening to a small residual oscillation of the BEC inside the trap which causes Doppler broadening. Absorption images of the expanding BEC indeed show random velocity fluctuations with a standard deviation of about 3-4 mm/s in the axial direction. A simple model of a damped harmonic oscillator driven by statistical fluctuations of the axial trap position can quantitatively explain these observations (see appendix 6.B).

Polarizability and ac-Stark shift

The main uncertainty in the calculations of the polarizability in chapter 6 is due to approximations made in estimating the contribution to the polarizability due to coupling to the ionization continuum. Since this contribution is far offresonant, we can neglect its wavelength dependence and treat it as a dc-offset. The calculated polarizabilities are given in atomic units which can be converted into SI using $\begin{bmatrix} 1 & a_0^3 \end{bmatrix}_{a.u.} = \begin{bmatrix} 4\pi\epsilon_0 a_0^3 \end{bmatrix}_{SI} \approx 1.64877 \times 10^{-41} \text{JV}^{-2}\text{m}^2$. The intensity of the laser beam can now be calculated from the scale of the polarizability compared to the ac-Stark shift using $I = 2\epsilon_0 ch\Delta\nu/\text{Re}(\Delta\alpha)$, where $\Delta\alpha$ is the differential polarizability, and $\Delta\nu$ is the observed ac-Stark shift [108].

Absolute frequency determination

The ODT laser wavelength is derived from both the spectroscopy laser and a second free-running fiber laser. Because the spectroscopy laser frequency is determined with much higher accuracy, the uncertainty on the ODT laser wavelength is dominated by the free-running laser. This wavelength is measured continuously during the course of the experiment using a high resolution wavemeter (High Finesse WSU-30) with a specified accuracy of 30 MHz, which was periodically calibrated on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{3}P_{2}$ line at 1083 nm. The laser wavelength was manually adjusted using the temperature control whenever it drifted by more than 50 MHz from the wavelength setpoint for that measurement week.

The spectroscopy laser is locked to an ultrastable laser at 1542 nm (Menlo systems) via an optical frequency comb to bridge the wavelength gap in a transfer-lock configuration [63]. The ultrastable laser serves as stable short term flywheel oscillator for the measurement. Over the course of a measurement day, the frequency of this reference is measured with respect to the Cs clock.

In order to reconstruct the absolute frequencies of the lasers several beatnotes are continuously measured with a zero-dead time frequency counter, referenced to the Cs clock. The frequencies which are measured are the frequency comb carrier offset frequency, the down-mixed pulse repetition rate, the spectroscopy laser beat-note (before mixing in the DDS), and either the virtual beat-note or the ultrastable laser beat note. The wavelengths of the lasers were measured using a wavemeter with sufficient resolution to determine the comb modenumber of the observed beat notes.

From these data the ultrastable laser frequency was reconstructed, and a linear fit allows us to compensate for the slow drift of this laser during the day. This drift was found to be 22(2) mHz/s $(1.1(1) \times 10^{-16} \text{ s}^{-1})$ on average and fluctuating from day to day with a standard deviation of 9 mHz/s. The modified Allan deviation of these data agrees well with the specified stability of the Cs clock at the measured time scales, typically reaching a stability in the low 10^{-13} region after a single measurement day. In total, the spectroscopy data were acquired over about 30 separate measurement days, yielding in total about 5×10^5 seconds of total integration time, which is enough to reach the clock's stability floor of 5×10^{-14} .

During the full measurement campaign the time delay between the Cs clock and GPS pulse per second signal was continuously measured. The Allan deviation of this delay averages down as τ^{-1} , and catches up with the Cs clock stability after about 10⁶ s. Integrating over the full course of the measurement campaign, which took several months (~ 8×10^6 s), we observed a fractional frequency drift of $-1.9(2) \times 10^{-13}$, with an accuracy that exceeds the specified Cs clock stability. We corrected for this drift in the frequency measurement data but take the specified clock stability floor as a conservative estimate of the uncertainty. The deviations between the GPS disseminated second with respect to the SI definition as reported in the BIPM circular T bulletin [124] were found to be negligible at the current level of uncertainty.

Data processing and statistical analysis

The measured transition frequency data are fit with a weighted linear least squares model regressing to the trap and probe laser powers (measured before and after each scan) and the chemical potential of the BEC (as determined from the MCP time-of-flight profile). In order to separate the purely statistical error from the error due to the systematic shifts, we calculate the point of minimum uncertainty from the covariance matrix of the fit. At this point the uncertainty on the transition frequency is not correlated to the uncertainty in the other parameters and can be considered purely statistical, amounting to 32 Hz. Extrapolating from this point to zero laser power and chemical potential is associated with a systematic uncertainty of 192 Hz, which constitutes the bulk of the uncertainty in this work.

6.A Appendix: Minor systematic effects

Vector and tensor polarizability

The measurement of the magic wavelength is more complicated when the tensor and vector polarizability are considered. The measured magic wavelength then depends on the details of the magnetic substates that are measured, on the polarization of the laser light and the orientation of the quantization axis. However, the tensor and vector polarizabilities are sufficiently small that these details do not affect the measured values at the current level of uncertainty.

First of all, the vector and tensor polarizabilities do not affect the frequency metrology in any way. The tensor polarizability drops out as common mode because we exclusively use the $m_J = \pm 1$ magnetic substates which are affected identically. The vector polarizability introduces an asymmetry between the $m_J = +1$ and $m_J = -1$ substates in the presence of an excess of either σ^+ or σ^- polarization but this asymmetry extrapolates to zero in the trap ac-Stark shift regression that is performed. The non-scalar terms in the polarizability are therefore only relevant to the measurement of the magic wavelength.

In an extension to the magic wavelength calculation (see appendix 4.A of chapter 4) the values $0.09 a_0^3$ and $0.03 a_0^3$ are for the vector and tensor polarizabilities respectively. These numbers are small compared to the scalar polarizability of 189.3 a_0^3 , and the 0.5 a_0^3 uncertainty on the dc-offset on the

polarizability curve as found in the magic wavelength measurement. Moreover, the effects of both the vector and tensor polarizability tend to average out in the geometry of our trap. Nominally, the trap laser polarization is linear in both beams with the polarization of the returning beam orthogonal to the first. This will create a rapidly oscillating polarization gradient over the trap, so that any polarization dependent terms average out.

Mean-field lineshape

The lineshape due to the mean-field shift is given by equation 25 of ref. [129], and can be written as

$$S(\Delta\nu) = AN \frac{\Delta\nu}{\delta\nu_{max}^2} \sqrt{1 - \frac{\Delta\nu}{\delta\nu_{max}}},$$
(6.2)

where A is a pre-factor not important to the discussion here, N is the number of atoms in the BEC, and $\Delta \nu$ is the detuning from resonance. The profile is equal to zero outside the interval $[0, \delta \nu_{max}]$. The profile's full width is given by

$$\delta\nu_{max} = \frac{4\pi\hbar^2(a_{ts} - a_{tt})}{hm}n(0) = \frac{a_{ts} - a_{tt}}{ha_{tt}}\mu,$$
(6.3)

where a_{tt} and a_{ts} are the triplet-triplet and triplet-singlet scattering lengths respectively, n(0) is the peak density of the BEC, and μ is the chemical potential of the BEC. A number of factors of two are different between equation 6.2 and equation 25 of ref. [129] because we use a one-photon, rather than a two-photon transition.

The average frequency shift of the lineshape can be calculated according to

$$\langle \Delta \nu \rangle = \frac{\int_0^{\delta \nu_{max}} \Delta \nu S(\Delta \nu) d\Delta \nu}{\int_0^{\delta \nu_{max}} S(\Delta \nu) d\Delta \nu}.$$
(6.4)

The two integrals can be solved using the simple substitution $x = \frac{\Delta \nu}{\delta \nu_{max}}$:

$$\int_{0}^{\delta\nu_{max}} \Delta\nu S(\Delta\nu) d\Delta\nu = \frac{AN}{\delta\nu_{max}} \int_{0}^{1} x^2 \sqrt{1-x} dx = \frac{16AN}{105\delta\nu_{max}}$$
(6.5)

$$\int_0^{\delta\nu_{max}} S(\Delta\nu) d\Delta\nu = \frac{AN}{\delta\nu_{max}^2} \int_0^1 x\sqrt{1-x} dx = \frac{4AN}{15\delta\nu_{max}^2}.$$
 (6.6)

We plug these expressions back into equation 6.4 to find

$$\langle \Delta \nu \rangle = \frac{4}{7} \delta \nu_{max} \tag{6.7}$$

Figure 6.5 shows the lineshape according to equation 6.2, along with the average shift according to equation 6.7.

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Figure 6.5: Lineshape of a BEC as a result of mean-field interactions according to equation 6.2. The average shift of the lineshape is analytically found to be 4/7 of the full width. The orange line indicates the same line profile with a 50% smaller chemical potential, giving rise to an observed shift of Δf_{MFS} .

Dc-Stark shift and blackbody radiation shift

The dc-Stark shift is given by [53]:

$$\Delta\nu = -\frac{1}{2h}\left\langle \vec{p} \cdot \vec{E} \right\rangle = -\frac{1}{2h} \operatorname{Re}(\Delta\alpha) E^2, \qquad (6.8)$$

where \vec{p} is the instantenous dipole moment, and \vec{E} the electric field. We know the difference in dc-polarizability between the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ states quite well: Re($\Delta \alpha$) = [483.55 a_{0}^{3}]_{*a.u.*} $\approx 7.97 \times 10^{-39}$ J V⁻² m² [169]. This gives a dc-Stark shift of

$$\frac{\Delta\nu}{E^2} = \frac{4\pi\epsilon_0 \times 483.55 \ a_0^3}{2h} \approx 6.02 \times 10^{-6} \ \text{Hz V}^{-2} \ \text{m}^2.$$
(6.9)

Based on a simulation of the electric field in the vacuum chamber using Comsol Multiphysics, we estimate that the ion-MCP produces a field of about 30 V/m at the position of the atoms. Even if we scale this up by a factor ten to be conservative the shift is smaller than 1 Hz.

This analysis also applies for the blackbody radiation (BBR) shift. The mean squared electric field from a blackbody is $\langle E^2 \rangle_T = (831.9 \text{ V m}^{-1})^2 \times \left(\frac{T}{300 \text{ K}}\right)^4$ [142], which gives a BBR shift of 4.17 Hz at 300 K. This estimate does not include the so-called dynamic contribution but this is typically smaller than 10% [181].

Quantum interference

Quantum mechanically, when multiple paths are available to reach the same final state, the question of which path was taken is ill-defined. In spectroscopy this can lead to frequency shifts due to off-resonant excitation of distant transitions. This is commonly referred to as "quantum interference" and was realized to be a significant problem for precision spectroscopy on the $2^{3}S \rightarrow 2^{3}P$ transitions [182].

In our work, we observe a transition between the 2 ${}^{3}S$ and 2 ${}^{1}S$ states. The latter has a natural lifetime of 20 ms in which it decays to the 1 ${}^{1}S$ ground state by the emission of two photons. Because we measure (via Penning ionization) the generated population of 2 ${}^{1}S$ atoms, quantum interference may occur with transitions to levels that decay to this state. By far the most important of these is the 2 ${}^{1}P$ state which decays into the 2 ${}^{1}S$ state with a branching ratio of about 0.1%. To estimate a possible quantum interference shift, we apply the analysis of ref. [183], which directly maps to our system when $|1 {}^{1}S\rangle, |2 {}^{3}S\rangle, |2 {}^{1}S\rangle, |2 {}^{1}P\rangle \rightarrow |0\rangle, |1\rangle, |2\rangle, |3\rangle.$

In the limit of short interaction time compared to the Rabi frequency of the main transition ($\Omega_2 T \ll 1$), we find that according to equation 21 of ref. [183], the transition frequency is shifted by

$$S_0 = \frac{\gamma_2 \gamma_3 + |\Omega_3|^2}{4\omega_{32}},\tag{6.10}$$

where γ_i is the inverse lifetime, and Ω_i the Rabi frequency for level $|i\rangle$, and ω_{ij} is the angular frequency difference between levels $|i\rangle$ and $|j\rangle$. It is only the first term in the numerator that corresponds to quantum interference, the second term is simply the ac-Stark shift from this particular level, which is already corrected for by the ac-Stark shift extrapolation.

 γ_3 and ω_{32} are known to be $2\pi \times 287$ MHz and $2\pi \times 146$ THz respectively [22]. For γ_2 there is some ambiguity: the natural linewidth is $2\pi \times 8$ Hz but the lifetime of this state is severely reduced because of Penning ionizing collisions with the remaining 2 3S atoms. The inverse of this lifetime however, cannot exceed the observed linewidth, indicating that $\gamma_2 < 2\pi \times 10$ kHz. In the interest of providing a conservative estimate, we will use this latter value since it gives the largest shift.

Evaluating equation 6.10 with these numbers gives a shift of approximately $2\pi \times 4.9$ mHz. However, the asumption $\Omega_2 T \ll 1$ is not justified, and a more thorough calculation is needed. We calculate the Rabi frequency according to

$$|\Omega_2|^2 = \frac{6\pi c^2}{\hbar\omega_{12}^3} A_{12} |\langle j_1 m_1 j_2 m_2 | JM \rangle|^2 I_0, \qquad (6.11)$$

where $\omega_{12} \approx 2\pi \times 192.5$ THz is the transition frequency, $I_0 \approx 5.5 \times 10^5$ W m⁻² is the peak intensity of the laser light, $A_{12} = 9.1 \times 10^{-8}$ s⁻¹ is the Einstein A-coefficient, and $\langle j_1 m_1 j_2 m_2 | JM \rangle \leq 1/\sqrt{3}$, depending on the polarization of the laser light, is the Clebsch-Gordan coefficient of the transition. From these numbers we calculate a Rabi-frequency that we round up to $2\pi \times 100$ Hz. Using this number we calculate the expected line profile from equation 25 of ref. [183], which has a slightly larger quantum interference shift of $2\pi \times 79.6$ mHz. This number should be understood as an absolute upper bound, the true shift is likely to be orders of magnitude smaller. For reference, table 6.2 shows all the parameters used to arrive at this estimate.

Table 6.2: Parameters used to calculate the upper bound estimate shift due to quantum interference. We take Ω_3 as a multiple of Ω_2 according to $\Omega_3/\Omega_2 = \sqrt{A_{13}/A_{12}}$ since they are derived from the same laser beam. Using these parameters, we find a shift of $\sim 2\pi \times 80$ mHz.

Parameter	value	Remarks
Ω_2	$2\pi \times 100 \text{ Hz}$	Upper bound
Ω_3/Ω_2	4×10^3	Same laser beam
γ_2	$2\pi \times 10 \text{ kHz}$	Upper bound
$\gamma_{2 \to 0}$	$2\pi \times 8 \text{ Hz}$	
γ_3	$2\pi \times 287 \text{ MHz}$	
ω_{23}	$2\pi \times 146 \text{ THz}$	
Т	$100 \mathrm{ms}$	

Second-order Zeeman shift

The second-order Zeeman shift of the 2 ${}^{1}S_{0}$ and 2 ${}^{3}S_{1}$ levels are given in [184] at 3.2 mHz G⁻² and 2.3 mHz G⁻² respectively. The ambient magnetic fields present during spectroscopy (typically about 0.5 G) would thus produce a shift < 0.3 mHz, which is completely negligible.

6.B Appendix: BEC oscillation and Doppler broadening

The random axial motion of the BEC inside the optical dipole trap mentioned in section 6.5 can be observed in a number of ways. The most straightforward of these is to measure the velocity of the expanding BEC after it is released from the trap using absorption imaging as shown in figure 6.6. Because the motion is stochastic, this velocity is uncorrelated between different experimental cycles but the root-mean-square (RMS) of the velocity is still well-defined. The magnitude of the velocity is found to be of the order of a few



Figure 6.6: Root-mean-square position fluctuations (with the average position subtracted) of the expanding BEC after release from the ODT as a function of expansion time. Each datapoint is averaged over 50 independent experimental cycles and the errorbar indicates the standard error of the mean. The fit indicates an RMS velocity of 2.92 mm/s but higher velocities have been observed on other days.

mm/s. Because the BEC is trapped, the atoms will be oscillating at the trap frequency with a positional amplitude of

$$\Delta x = \Delta v / \omega_0 = (4 \text{ mm/s}) / (2\pi \times 27 \text{Hz}) \approx 24 \mu \text{m}$$
(6.12)

which is fairly small compared to the size of the BEC $R = \sqrt{2\mu/m\omega_0^2} \approx 190 \ \mu \text{m}$ (assuming a typical value for the chemical potentical $\mu = k_B \times 0.3 \ \mu \text{K}$).

This velocity is sufficient to generate a Doppler shift of several kHz ($\Delta \nu_{Dopp} = v/\lambda = (4 \text{ mm/s})/(1.557 \,\mu\text{m}) \approx 2.6 \text{ kHz}$) which is large enough to have an influence on the spectroscopy signal. One effect of this is that the time-dependent excitation signal (figure 6.7) contains a modulation at both the axial trap frequency and at double this frequency, with relative strengths depending on detuning. A heuristic explanation of this behaviour is that if the laser were exactly resonant without the oscillation the oscillation brings it in resonance when the BEC is standing still, which occurs twice per oscillation cycle. If however, the laser would be just outside of resonance without the oscillation, some specific velocity will bring it into resonance which occurs only once per oscillation cycle.

It is not directly obvious how this oscillation affects the spectroscopy lineshape. In order to better understand what is happening I model the trap as a



Figure 6.7: Time-dependent ion-MCP signal during excitation. The amplitude of the signal is modulated with a frequency dependent on the detuning. On resonance (left), the modulation frequency is the double of the trap frequency (~ 30 Hz) while slightly off-resonance (right) the frequency is equal to the trap frequency.



Figure 6.8: Measurement of the axial trap frequency and damping rate using absorption imaging. An oscillation is induced using a magnetic field gradient after which the BEC is held inside the ODT for a certain hold time. The datapoints indicate the center position of the BEC after 20 ms of expansion time. The BEC undergoes damped harmonic oscillation inside the trap. From an exponentially damped sinusoidal fit (red line) the trap frequency $\omega_0 = 2\pi \times 27$ Hz and damping rate 6 s⁻¹ are derived.



Figure 6.9: Simulations of the motion of the BEC inside the trap and its effect on the spectroscopy signal. Left: Time-dependent velocity of the BEC in the trap subjected to 1 μ m/ \sqrt{Hz} white noise up to 100 Hz. The RMS velocity is about 4 mm/s. Right: Simulated ion-MCP signal with velocity noise, assuming a 5 kHz laser linewdith. The traces from top to bottom indicate the signal for the same velocity trace but different detuning: Green, +6 kHz; Orange, +3 kHz; Blue, on resonance; Red, -3 kHz; Purple, -6 kHz.

damped harmonic oscillator with the differential equation [185]

$$\ddot{x} + 2\gamma \dot{x} + \omega_0^2 x = \frac{1}{m} F(t).$$
(6.13)

The damping rate $\gamma = 6 \text{ s}^{-1}$ and eigenfrequency $\omega_0 = 2\pi \times 27$ Hz can be measured using absorption imaging as shown in figure 6.8. This damping is attributed to collisions with the thermal fraction of the gas and is of comparable magnitude to the damping rate found in other experiments [186]. Using equation 6.13 the motion of the BEC subject to a force can be simulated. This force is postulated to arise from random fluctuations in the axial trap position. A deviation of the trap position exerts a force on the BEC given by

$$F = -m\omega_0^2 \Delta x. \tag{6.14}$$

Plugging this into equation 6.13 and numerically solving for a white noise input allows simulation of the movement of the BEC. The result is shown in figure 6.9. A noise magnitude of $1 \ \mu m/\sqrt{\text{Hz}}$ in a 100 Hz bandwidth was chosen to produce about 4 mm/s RMS velocity fluctuations (slightly higher but similar to what is observed in absorption imaging).

The time-dependent ion-MCP signal corresponding to this motion is given by the laser profile with a time-dependent detuning

$$S(t,\Delta f) \propto \frac{1}{(\Gamma/2)^2 + (\Delta f + v(t)/\lambda)^2}.$$
(6.15)

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Figure 6.10: Simulated line profile for a randomly oscillating BEC. The black points are the integrated signal plus a small Gaussian noise term at different detunings of independent simulations of BEC motion. These simulations assume $1 \,\mu m/\sqrt{\text{Hz}}$ position noise and use the measured trap frequency and damping rate. The blue dashed line is a Lorentzian profile with a 5 kHz FWHM corresponding to the unmodulated laser profile, and the red dashed line is a Gausian fit to the simulated datapoints.

Figure 6.9 shows the simulated ion-MCP signal using this formula, showing qualitative agreement with the observed signals (figure 6.7).

The lineshape observed in spectroscopy is obtained simply by integrating the time signal $S(t, \Delta)$ over the probe time. For a 100 ms probe time these lineshapes will be quite erratic because this probe time is comparable to the oscillation time (~ 40 ms). This results in frequency noise on the lineshape which has a magnitude given by the Doppler shift for the mean velocity during the excitation. This number can be found by taking the total distance travelled divided by the probe time i.e. $(\Delta x/T)/\lambda = (27 \ \mu m/100 \ ms)/1.557 \approx 0.2 \ kHz$. Figure 6.10 shows a simulated spectroscopy line profile, which is qualitatively similar to the observed signals and reproduces the 10 kHz linewidth.

In order to reproduce the observed velocity fluctuations a noise on the axial trap position of 1 $\mu m/\sqrt{\text{Hz}}$ is required which is small compared to the beam waist of the trap laser. Because of the trap geometry involving refocussing lenses any beam pointing noise on the incoming trap beam is to first-order compensated by the returning beam. The most likely source for fluctuations is therefore beam pointing noise unique to the returning beam. At the time of writing it has not yet been verified that this noise is of the required magnitude.

Chapter 7

Conclusions and Outlook

The main goal of the work presented in this thesis was to substantially improve the previous measurement of the isotope shift on the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition in order to determine the nuclear charge radius difference. The improved measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency in ⁴He reported in chapter 6 is an important step in the pursuit of this goal. The measured value disagrees slightly with the value measured by van Rooij *et al.* [21], at a significance of 1.7 σ , as shown in figure 7.1. As was mentioned in chapter 6, the discrepancy may be caused by their estimate of the mean field shift which is slightly too small compared to the figure found in this work.

The final step, an improved measurement of the ³He transition frequency, is currently underway. For now, using the ³He measurement from van Rooij *et al.*, an updated value for the squared nuclear charge radius difference of $\delta r^2 = 1.041(7)$ fm² can be found (see figure 1.3 for a comparison with other measurements and table 7.1 for details of the calculation). Despite a small shift



Figure 7.1: Comparison of the two measurements of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency in ⁴He. RvR11 and Reng18 indicate, respectively, van Rooij *et al.* [21] and this work (chapter 6).

Table 7.1: Calculation of the squared nuclear charge radius difference. The calculation is performed according to Pachucki *et al.* [31].

Term	Value	Ref.
$+2 \ {}^{3}S_{1}^{F=3/2} \rightarrow 2 \ {}^{1}S_{0}^{F=1/2} \ ({}^{3}\text{He})$	$+192\ 504\ 914\ 426.4(1.5)\ \mathrm{kHz}$	[21]
$-2 {}^{3}S_{1} \rightarrow 2 {}^{1}S_{0} (^{4}\text{He})$	-192 510 702 148.72(20) kHz	Chap. 6
$+2 {}^{3}S_{1}^{F=3/2}$ hyperfine splitting	-2 246 567.056(5) kHz	[85]
$-2 {}^{3}S \rightarrow 2 {}^{1}S$ (point nucleus)	$+8\ 034\ 065.91(19)\ \mathrm{kHz}$	[31]
Total nuclear size shift	-223.47(1.5) kHz	
\div Proportionality constant	\div -214.66(2) kHz/fm ²	[31]
$\delta r^2 (^3 \text{He} - {}^4 \text{He})$	$1.041(7) \text{ fm}^2$	

in the direction of the result of Shiner [84], the improved accuracy means that the discrepancy still stands at 2.6σ . The uncertainty on this updated value is completely determined by the uncertainty on the ³He frequency however, and if a new measurement can be performed with similar accuracy to what was attained in ⁴He, the nuclear charge radius difference can be determined with an accuracy better than 0.002 fm².

Future work

If this measurement can indeed be realized, the experimental accuracy on the isotope shift will not be far behind the current uncertainty of the theory value. Recent theoretical work is already anticipating significant improvement however, indicating that further improvements to the experimental accuracy can advance the resolution on the nuclear charge radius still further. With the experience gathered in this work a number of technical modifications can be suggested that may yet yield substantially better experimental accuracy.

Since the most important systematic uncertainties found in chapter 6 can still be reduced with more accurate measurements, improvements in the observed linewidth should be considered the first line of attack. The Doppler shift due to trap oscillations described in section 6.B should be investigated further. If, as is suggested, these oscillations are indeed driven by fluctuations in the axial trap position they may be compensated by actively stabilizing the beam pointing of the trap laser beams. Reducing the laser linewidth further is also of importance. As proposed by Notermans [87], this can be achieved by actively compensating the phase noise introduced by the fiberlink between the frequency-comb lab and the He^{*} setup (and to a lesser extent the fiberlink between the frequency comb and the ultrastable laser) and improving the lock electronics. Finally, stabilizing the magnetic field in the science chamber of the He^{*} setup might make the experiment still less dependent on drifts of the ambient magnetic field in the lab. As discussed in section 2.3, further improvement may be gained by employing an optical lattice at the magic wavelength. The extra confinement of the lattice would strongly alter the density of states, allowing spectroscopy to be done in the Lamb-Dicke regime. This would produce a sharp reduction in the linewidth of the ³He^{*} degenerate Fermi gas, but it would also allow measurements in ⁴He at lower density, possibly in a thermal gas, to reduce the mean field shift. The practical implementation of an optical lattice will be challenging however, because of the high intensity necessary to produce trap depths exceeding the recoil energy (see section 2.3). Barring an extraordinary increase in the available laser power, the only ways to produce the intensities required would be to focus very tightly or to use a resonant build-up cavity.

With these technical modifications the experimental accuracy on the absolute transition frequencies may be further improved. Considering the small magnitude of various systematic effects estimated in section 6.A such a measurement would mainly be limited by the stability of the Cs-clock (5×10^{-14}), i.e. a factor 20 improvement over the result of chapter 6. This limitation is also technical however, and may be overcome with a better clock or by measuring the isotope shift directly instead of comparing absolute transition frequencies. Ultimately there is no reason to suppose that with considerable effort the absolute 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency can not be measured to an accuracy rivalling those of state-of-the-art optical lattice clocks.

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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, R.J. Rengelink, & W. Vassen. Comparison of spectral linewidths for quantum degenerate bosons and fermions. *Phys. Rev. Lett.* 117, 213001 (2016)
- **Chapter 4:** R.P.M.J.W. Notermans, R.J. Rengelink, K.A.H. van Leeuwen & W. Vassen. Magic wavelengths for the $2 \ {}^{3}S \rightarrow 2 \ {}^{1}S$ transition in helium. *Phys. Rev. A* **90**, 052508 (2014)
- Chapter 5: R.J. Rengelink, R.P.M.J.W. Notermans & 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)
- Chapter 6: R.J. Rengelink, R.P.M.J.W. Notermans, Y. van der Werf, R. Jannin, K.S.E. Eikema, M.D. Hoogerland & W. Vassen. Precision spectroscopy of helium in a magic wavelength optical dipole trap. *Nature Physics* Published online doi:10.1038/s41567-018-0242-5 (2018)

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- (i) 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.
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- (iii) J.J. Renema, R.J. Rengelink, I. Komen, Q. Wang, R. Gaudio, K.P.M. op 't Hoog, Z. Zhou, D. Sahin, A. Fiore, P. Kes, J. Aarts, M.P. van Exter & M. de Dood. The effect of magnetic field on the intrinsic detection efficiency of superconducting single-photon detectors. *Appl. Phys. Let.*, **106**, 092602 (2015)

Summary

Named after $H\lambda \iota o \varsigma$, the greek titan god of the sun, the helium atom has played a pivotal role in the development of modern physics. As a two-electron atom, the study of the electronic structure of helium has allowed physicists to generalize the model of the well-understood one-electron system (i.e. the hydrogen atom) to the rest of the periodic table. To this day, the most important experimental method to study atomic physics is spectroscopy, the precise measurement of the frequencies corresponding to the wavelengths of light emitted or absorbed by the object under study. For the helium atom, this light is emitted at a number of discrete frequencies which are predicted to exceptional precision by the theory of quantum electrodynamics (QED). QED is the besttested constituent of the standard model of physics, the consistent unification of all forces of nature except gravity (which does not play a meaningful role at the length scale of atoms). The non-intuitive quantum effects predicted by this theory produce minute frequency shifts and by performing equally precise measurements these effects can be experimentally verified.

Despite the enormous success of QED in explaining phenomena in atomic physics and beyond, a recent problem found in the hydrogen atom may indicate that it is not the full story. This problem arose when researchers sought to measure the frequency shift of a spectral line in hydrogen due to the very small vet finite size of its nucleus, i.e. the proton. Assuming that QED is correct this would allow the size of the proton to be determined experimentally to very high accuracy. These measurements were performed both in regular hydrogen and also in a variant, muonic hydrogen, wherein the electron has been replaced by an elementary particle called a muon. The muon is identical to the electron in every way except for its mass which is 200 times higher. Because of this higher mass, the muon orbits much closer to the nucleus so that the finite nuclear size effect is amplified and easier to measure. Surprisingly, the size of the proton as measured using muonic hydrogen was found to be about 4% smaller than for regular hydrogen at a high statistical significance. This finding has since become known as "The proton size puzzle" and, provided no error was made in the execution and interpretation of these measurements, is evidence of a hitherto unknown interaction between fundamental particles indicating that the standard model is incomplete.

Ultracold metastable helium

Hoping to gain more insight into this problem, researchers have also shifted their attention to muonic helium. The primary motivation for the work in this thesis is to contribute to this line of research by providing a very accurate reference measurement in regular helium to compare the results in muonic helium to. Performing spectroscopy on helium in the ground state is technically very difficult due to the required wavelengths which are in the extreme ultraviolet region of the electromagnetic spectrum. Instead spectroscopy is performed on the metastable 2 ${}^{3}S_{1}$ state. Even though this state is very highly excited (its internal energy is 19.82 eV, enough to ionize any other atom except neon) and will therefore eventually decay into the ground state, it is very long-lived (8000 seconds) and has a transition to the 2 ${}^{1}S_{0}$ state at the near infrared wavelength of 1557 nm. This transition is sensitive to the nuclear finite size and is very narrow (in technical terms, it has a very high quality factor of 2.5×10^{13}) which allows for very accurate measurements.

A precision measurement of the helium nucleus can then be performed by very accurately measuring the difference of this transition frequency between helium atoms which have a nucleus containing two protons and two neutrons (⁴He, the vast majority of all helium found on earth) and those with a nucleus containing two protons and a single neutron (³He, roughly one atom for every 100,000 ⁴He atoms found on earth). A downside however is that the transition breaks two electric dipole selection rules so that we speak of a doubly-forbidden transition. While forbidden in this case does not mean impossible, the transition is more than a quadrillion (10¹⁵) times weaker than the normal dipole-allowed transitions that are typically observed in the helium spectrum.

In order to be able to drive this extremely weak transition more easily, the helium gas is cooled down to less than one-millionth of a degree above absolute zero (-273.15°C) using laser cooling techniques. At such low temperatures the thermodynamic behaviour of the gas is driven by quantum statistics which depend strongly on a fundamental binary property of the particles that make up the gas; a particle can either be a boson (integer spin) or a fermion (half-integer spin). In chapter 3 the difference in quantum statistics depending on this property is observed to lead to a dramatic difference in the absorption lineshape found for ⁴He (a boson), and ³He (a fermion). These lineshapes are in good agreement with existing theoretical models when the effects of the so-called ac-Stark shift (see next section) are taken into account.

A magic wavelength optical dipole trap

To keep the atoms at extremely low temperature they must remain isolated from the environment in some sort of trap that does not heat the atoms up again too quickly. The type of trap used in this work is called an optical dipole trap and it consists of an intense focused laser beam that produces an energy shift of the atoms known as the ac-Stark shift. When the wavelength of this laser is properly chosen the shift is strongly negative so that the laser beam forms a potential well and the atoms experience a pulling force when they move out of the center of the laser beam. In the previously most accurate measurement of the 2 ${}^{3}S_{1} \rightarrow 2 {}^{1}S_{0}$ transition frequency the optical dipole trap was an important source of experimental uncertainty. Although the atoms in the 2 ${}^{3}S_{1}$ state experienced a strong negative energy shift, this shift was strongly positive for the excited 2 ${}^{1}S_{0}$ state, producing a large systematic frequency shift which had to be corrected for.

Most of the work described in this thesis has been to implement a so-called magic wavelength trap which is a remedy to exactly this problem. The sign and magnitude of the ac-Stark shift depend on the polarizability of an atomic state and for certain wavelengths (known as *magic wavelengths*) the polarizabilities of two states are exactly the same. In an optical dipole trap at that wavelength both states experience exactly the same potential irrespective of the intensity or spatial profile of the laserlight. For the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ states the wavelength dependent polarizabilities are calculated in chapter 4 allowing a number of magic wavelengths to be identified. The most promising of these is found in the ultraviolet part of the electromagnetic spectrum at 319.815 nm for ⁴He. In order to generate an optical dipole trap at this wavelength a powerful laser system was built which is the subject of chapter 5.

With this new laser system as well as a number of other improvements, a new measurement was performed of the 2 ${}^{3}S \rightarrow 2 {}^{1}S_{0}$ transition in ⁴He which is discussed in chapter 6. Besides the transition frequency, this measurement allowed a very accurate determination of the magic wavelength which is in excellent agreement with the value calculated in chapter 4 as well as a more accurate calculation performed by another group since then. The theory developed in chapter 3 was used to model the energy shift caused by the interaction between the 2 ${}^{3}S_{1}$ and 2 ${}^{1}S_{0}$ atoms which allowed the determination of the scattering length characterizing low-temperature collisions between these states. Finally, the transition frequency measurement was improved by almost a factor ten, corresponding to a relative accuracy of 1.0×10^{-12} , the highest accuracy to which any optical transition in the helium atom has ever been measured up to now. If the distance of the moon to the earth were to be measured at the same accuracy the measurement error would be approximately equal to the thickness of the cover of this thesis.

Samenvatting

Het helium atoom, vernoemd naar Ἡλιος, de zonnegod uit de Griekse mythologie, heeft een cruciale rol gespeeld in de ontwikkeling van de moderne natuurkunde. Met zijn twee elektronen leidde de ontrafeling van de structuur van het helium atoom tot het begrip hoe de theorie van het één-elektron systeem (d.w.z. het waterstof atoom) gegeneraliseerd kon worden naar de rest van het periodiek systeem. Nog altijd is de spectroscopie, het nauwkeurig meten van de frequenties die overeenkomen met de golflengtes van licht die worden uitgezonden of geabsorbeerd door een object, de belangrijkste experimentele methode in de atoomfysica. In het geval van het helium atoom wordt dit licht enkel bij een aantal specifieke frequenties uitgezonden welke nauwkeurig voorspeld worden door de kwantumelektrodynamica (QED, van het Engelse quantum electrodynamics). De QED is het best geteste onderdeel van het standaardmodel van de natuurkunde, de consistente vereniging van alle bekende natuurkrachten behalve de zwaartekracht (welke geen rol van betekenis speelt op de lengteschaal van atomen). Door deze frequenties met eenzelfde hoge nauwkeurigheid te meten kunnen de zeer niet-intuïtieve kwantumeffecten die deze theorie voorspelt experimenteel worden bevestigd.

Hoewel de QED uitzonderlijk succesvol is geweest in het verklaren van fenomenen uit de atoomfysica en daarbuiten is er recent een probleem ontstaan in de spectroscopie van het waterstofatoom dat mogelijk niet volledig binnen de QED begrepen kan worden. Dit probleem ontstond toen onderzoekers de frequentieverschuiving van een spectraallijn in waterstof probeerden te meten die veroorzaakt wordt door de zeer geringe doch eindige grootte van de atoomkern oftewel het proton. Vanuit de veronderstelling dat QED volledig juist is zou deze grootte daarmee experimenteel met zeer hoge nauwkeurigheid bepaald kunnen worden. Deze metingen zijn uitgevoerd in normaal waterstof maar ook in een variant daarvan, muonisch waterstof, waarvan het elektron vervangen is door een elementair deeltje genaamd het muon. Het muon is identiek aan het elektron in elk opzicht met uitzondering van de rustmassa welke 200 maal groter is. Vanwege deze hogere massa beschrijft het muon een veel kleinere baan rondom de atoomkern wat de frequentieverschuiving door de eindige kerngrootte uitvergroot en daardoor makkelijker te meten maakt. Tegen alle verwachtingen in bleek de grootte van het proton bepaald in muonisch waterstof met hoge statistische significantie ongeveer 4% kleiner te zijn dan dezelfde waarde bepaald in normaal waterstof. Deze vondst staat bekend als de "protonstraalpuzzel" en, gegeven dat er geen fout is gemaakt bij de interpretatie of uitvoering van deze experimenten, vormt bewijs van een tot dusver onbekende wisselwerking tussen elementaire deeltjes wat aangeeft dat het standaardmodel incompleet is.

Ultrakoud metastabiel helium

In de hoop meer grip op dit probleem te krijgen hebben onderzoekers hun aandacht sindsdien ook naar het heliumatoom verlegd. De voornaamste motivatie voor het werk in dit proefschrift is om aan deze onderzoekslijn bij te dragen door een zeer nauwkeurige meting in het normale heliumatoom uit te voeren zodat deze vergeleken kan worden met muonisch helium. Spectroscopie van helium in de grondtoestand is technisch niet eenvoudig is vanwege de vereiste golflengten welke in het extreem ultraviolette deel van het elektromagnetisch liggen. In plaats daarvan wordt er gewerkt met de metastabiele 2 ${}^{3}S_{1}$ toestand. Hoewel deze toestand met een interne energie van 19.82 eV bijzonder hoog aangeslagen is en daarom uiteindelijk terug naar de grondtoestand zal vervallen, is de natuurlijke levensduur uitzonderlijk lang (8000 seconden) en kan vanuit deze toestand een overgang gemaakt worden naar de 2 ${}^{1}S_{0}$ toestand met een golflengte in het nabije infrarood van 1557 nm. Deze overgang is gevoelig voor het effect van de eindige kernstraal en heeft een zeer hoge kwaliteitsfactor (2.5×10^{13}) waardoor zeer nauwkeurige metingen gedaan kunnen worden.

Een precisiemeting van de heliumkern kan uitgevoerd worden door het verschil in deze overgangsfrequentie te meten tussen heliumatomen met een kern bestaande uit twee protonen en twee neutronen (⁴He, waaruit vrijwel al het aardse helium bestaat) en atomen met een kern bestaande uit twee protonen en slechts één neutron (³He, waarvan op aarde ruwweg één voor elke 100,000 ⁴He atomen gevonden wordt). Een nadeel van deze overgang is echter dat twee selectieregels voor elektrische dipool overgangen gebroken worden waardoor we over een dubbel-verboden overgang spreken. Hoewel verboden in dit geval niet gelijk staat aan onmogelijk is de overgang meer dan een biljard (10¹⁵) maal zwakker dan normale dipool-toegestane overgangen die typisch in het heliumspectrum gevonden worden.

Om deze extreem zwakke overgang iets gemakkelijker te kunnen maken wordt het heliumgas tot minder dan één miljoenste graad boven het absolute nulpunt (-273.15° C) afgekoeld met behulp van laserkoeltechnieken. Bij zulke lage temperaturen wordt het thermodynamisch gedrag van het gas bepaald door de kwantumstatistiek welke sterk afhangt van een fundamentele binaire eigenschap van de deeltjes waar het gas uit bestaat; een deeltje kan ofwel een boson zijn (de zogenaamde spin van het deeltje is een geheel getal), danwel een fermion (spin van het deeltje is de helft van een oneven getal). In hoofdstuk 3 wordt het grote verschil beschreven dat deze eigenschap veroorzaakt voor de kwantumstatistiek waardoor de vorm van de absorptielijn voor ⁴He (een boson) radicaal anders is dan voor ³He (een fermion). De geobserveerde lijnprofielen kunnen goed verklaard worden met bestaande theoretische modellen wanneer het effect van de zogenaamde dynamische Starkverschuiving (zie volgende paragraaf) op de juiste manier wordt meegenomen

Een optische dipoolval bij een magische golflengte

Om de atomen langere tijd op zeer lage temperatuur te houden is het nodig ze goed te isoleren van de veel warmere omgeving door middel van een "val" die niet teveel opwarming veroorzaakt. Het type val dat in dit werk gebruikt wordt heet een "optische dipoolval" en bestaat uit een intense gefocusseerde laserbundel welke een energieverschuiving bij de atomen verzoorzaakt die bekend staat als de dynamische Starkverschuiving. Bij een goed gekozen golflengte van de laser is deze verschuiving sterk negatief waardoor de laserbundel een potentiaalput vormt en de atomen een aantrekkende kracht ondergaan wanneer ze zich van de optische as van de laserbundel af bewegen. In de voorheen meest nauwkeurige meting van de 2 $^3S_1 \rightarrow 2 \, ^1S_0$ overgangsfrequentie was de gebruikte optische dipool val een belangrijke bron van experimentele onzekerheid. Hoewel de atomen in de 2 3S_1 toestand een sterke negatieve energieverschuiving voelden was deze juist sterk positief voor de aangeslagen 2 1S_0 toestand waardoor gecorrigeerd moest worden.

Het grootste deel van het werk beschreven in dit proefschrift is de implementatie van een zogenaamde magische golflengte val waarmee getracht wordt dit probleem op te lossen. Het teken en de grootte van de dynamische Starkverschuiving hangen af van de polarizeerbaarheid van een toestand en voor bepaalde golflengtes van licht (de zogeheten magische golflengtes) kunnen de polarizeerbaarheden van twee verschillende toestanden exact aan elkaar gelijk zijn zodat beide toestanden in een optische dipoolval bij deze golflengte gevangen kunnen worden in exact dezelfde potentiaal ongeacht de intensiteit of het ruimtelijk profiel van de laserbundel. Berekeningen van de polarizeerbaarheden van de 2 ${}^{3}S_{1}$ en 2 ${}^{1}S_{0}$ toestanden staan beschreven in hoofdstuk 4. Op basis van deze berekeningen worden een aantal magische golflengten gevonden waarvan de meest bruikbare gevonden wordt bij 319.815 nm voor ⁴He, in het ultraviolette deel van het elektromagnetisch spectrum. Om bij deze golflengte

een optische dipoolval te creëren is een krachtige laser gebouwd welke het onderwerp van hoofdstuk 5 vormt.

Met deze nieuwe laseropstelling, alsmede een aantal andere verbeteringen, is een nieuwe meting uitgevoerd van de 2 ${}^{3}S \rightarrow 2 {}^{1}S_{0}$ overgang in ⁴He, hetgeen beschreven staat in hoofdstuk 6. Naast de overgangsfrequentie leidde deze meting tot een zeer nauwkeurige bepaling van de magische golflengte welke in uitstekende overeenstemming is met de berekeningen in hoofdstuk 4 maar ook met meer nauwkeurige berekeningen die sindsdien door anderen gedaan zijn. De theorie van de absorptielijnvorm die ontwikkeld is in hoofdstuk 3 is gebruikt om de wisselwerking tussen de 2 ${}^{3}S_{1}$ en 2 ${}^{1}S_{0}$ atomen te beschrijven waardoor de zogeheten verstrooiingslengte, die botsingen tussen deze toestanden bij lage temperatuur beschrijft, bepaald kon worden. Ten slotte is de meting van de overgangsfrequentie met bijna een factor tien verbeterd tot een relatieve nauwkeurigheid van 10^{-12} , tot nog toe de meest nauwkeurige optische frequentiemeting in het heliumatoom. Wanneer de afstand van de aarde tot de maan met dezelfde nauwkeurigheid zou worden gemeten zou de meetfout overeenkomen met de dikte van de kaft van dit proefschrift.

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Allereerst zou ik elke promovendus een begeleider toewensen die zo voor je klaar staat als mijn begeleider Wim V dat voor mij heeft gedaan. Zijn deur stond altijd voor me open voor een gesprek over natuurkunde, of het nou direct met het experiment te maken had of niet. Uit de archiefkasten op zijn kantoor bleek altijd nog wel een interessant achtergrondartikeltje te komen of er was nog wel een mailtje van Drake of Pachucki dat zeer van pas kwam bij het schrijven van mijn proefschrift. Ook toen het nodig was om persoonlijke zaken prioriteit te geven heb ik mij daarin altijd zeer gesteund gevoeld. Dank daarvoor.

Lange tijd heeft Remy samen met mij op de metastabiel helium BEC opstelling gestaan. Ondanks ons gevoel voor humor is deze samenwerking zeer productief geweest getuige de mooie verzameling artikelen waarvan wij beiden auteur zijn. Ik zal altijd met net zo veel plezier terugdenken aan onze wetenschappelijke discussies als aan onze minder wetenschappelijke gesprekken onder het genot van een manke monnik.

Toen Remy naar Californië vertrok stond ik enige tijd alleen op de opstelling maar al gauw kreeg ik versterking van Yuri. Ik ben ervan onder de indruk hoe snel hij zich het experiment eigen heeft weten te maken waardoor hij uiteindelijk de laatste metingen van mij over heeft kunnen nemen. Rafaël joined the team while I was already writing but nevertheless proved instrumental in understanding the final systematic effects. Ook Maarten leverde tijdens zijn sabbatical een bijdrage aan de frequentiemeting. Niet alleen vanwege zijn uitgebreide kennis en ervaring maar ook omdat dankzij zijn PR-inspanningen onze meting tot in Nieuw-Zeeland de kranten heeft gehaald.

Als je nog eens wat op wil steken van je promotieonderzoek is het geen slecht idee eens met Kjeld door het frequentiekamlab te lopen. Ook hem wil ik bedanken, niet alleen heeft hij zijn frequentiekam en ultrastabiele laser ter beschikking gesteld, hij heeft ook mijn analyses en manuscripten van scherpe

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kritieken en commentaar voorzien. Wanneer je Kjeld er eenmaal van hebt kunnen overtuigen dat je meting goed in elkaar zit heb je geen refereecommentaar meer te vrezen.

During a crucial period of the final measurement campaign it was necessary to share the comb with Sayan and Matthias of the HD⁺ experiment, Cunfeng and Joël of the H₂ experiment, and Frank of the NICE-OHMS experiment. Despite having to share we were all able to use the comb effectively by synchronizing our measurements and cleverly choosing comb rep rates. The data taken during this period have led to no less than three high-profile publications. I believe we can all be extremely proud of this achievement which is in no small part thanks to our strong ability to cooperate.

Ook de andere leden van de metastabiel helium groep mogen uiteraard niet ontbreken. Toen ik begon als promovendus begon Ruud als masterstudent en bleef daarna ook aan als promovendus. Derhalve was Ruud een constante factor tijdens mijn promotietraject en een hele aimabele en collegiale factor bovendien. Wanneer het op laserkoelen en koude atomen aankwam kon je van niemand in het lab meer leren dan van Steven. Zijn overstap naar het KNMI is een groot verlies voor de atoomfysica geweest. I very much enjoyed my conversations with Hari and Adonis, as well as the excellent indian lunches Hari used to bring to work.

Wanneer er in de aml-groep iets gemaakt moet worden is de technicus Rob altijd de eerste en belangrijkste verdedigingslinie. Dat de opstelling ondanks een aantal stroomuitvallen, lekkages en van ouderdom gesneuvelde apparatuur goed is blijven werken valt grotendeels op zijn conto te schrijven. Ook de mensen van de mechanische en elektronische werkplaats hebben bijgedragen aan het goed (blijven) functioneren van de opstelling. In het bijzonder wil ik Tim Kortekaas noemen voor zijn hulp bij de transfer-lock opstelling.

Ik heb mij ruim vier jaar lang zeer thuis gevoeld in de aml-groep onder leiding van mijn copromotor Wim U. Met name uit gesprekken met Edcel, Jeroen, die er genoegen in leek te scheppen obscure systematische effecten te verzinnen en te vragen of we daar al aan hadden gedacht, Tjeerd, Matthijs, Stefan en Chantal heb ik veel op kunnen steken. Daarnaast was er ook veel gezelligheid tijdens kerstborrels en barbeques, potjes schaak, rondjes schaatsen, spare-ribs eten, of gewoon borrelen met de leden van de groep. Deze prettige sfeer is te danken aan Aernout, Allan, Anne, Charlène, Itan, Jurriaan, Laura, Luca, Marina, Mario, Nicoletta, Rob, Robert, Vasco, en vele anderen.

Naast de collega's aan de VU wil ik ook een aantal collega's van andere universiteiten noemen. Arthur, Alex, Alexander, Atreju, Bas, Benny, Daniël, Chris, Elwin, Erik, François, Flavio, Frank, Jacco, Julian, Lydia, Maarten, Maurits, Nivedya, Olivier, Roeland, Sean, Qiang en natuurlijk Jelmer, die ik eeuwig teleurgesteld heb met mijn vertrek naar de Dark Side. Tot slot wil ik mijn familie bedanken voor alle steun, liefde en geduld. Mijn vader en moeder die altijd voor me klaar stonden, Andrea met wie ik al bijna tien jaar lief en leed gedeeld heb en die mij blijft inspireren het beste in mijzelf naar boven te halen, en ten slotte Elian die mij zelfs op mijn donkerste dagen nog kan laten glimlachen. Ik kijk ook heel erg uit naar de avonturen die we samen met kleine Floris gaan beleven maar dat is een verhaal voor een andere keer. Precision spectroscopy of light atomic systems allows the sizes of nuclei to be probed with high accuracy. Surprisingly, different experiments of this type in the hydrogen atom have yielded conflicting results, a problem that has since become known as **the proton radius puzzle**.

The work described in this thesis seeks to probe the size of the helium nucleus instead, using helium atoms in the metastable state. To achieve this goal, small atomic ensembles are laser cooled to less than a microkelvin above absolute zero and trapped in the focus of an intense laser beam. By carefully tuning the wavelength of the trapping laser to a so-called **magic wavelength**, the distorting influence of the trap laser on the spectroscopy measurement can be canceled. By performing ab initio calculations of the atomic polarizability, a promising magic wavelength is found in the ultraviolet part of the spectrum and a powerful laser system is constructed to operate a trap at this wavelength. Using this trap, the frequency of radiation required to drive an exceedingly weak transition is measured with extreme accuracy. Thus the single most accurate spectroscopic measurement in the helium atom to date is realized, probing the size of its nucleus at sub-attometer precision.